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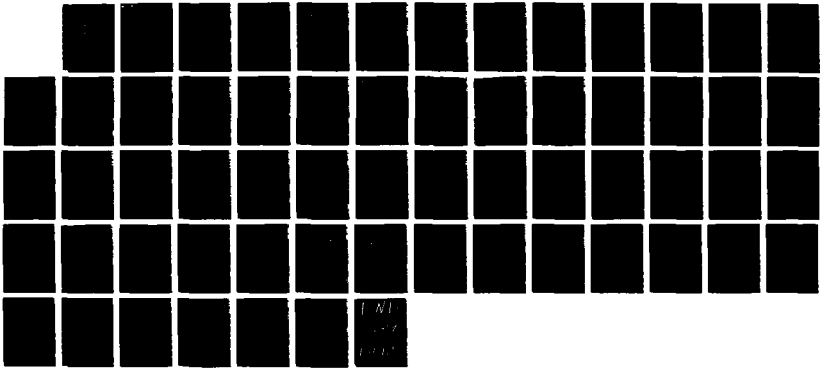

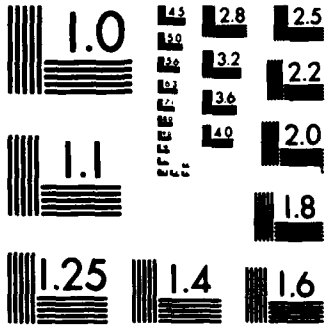

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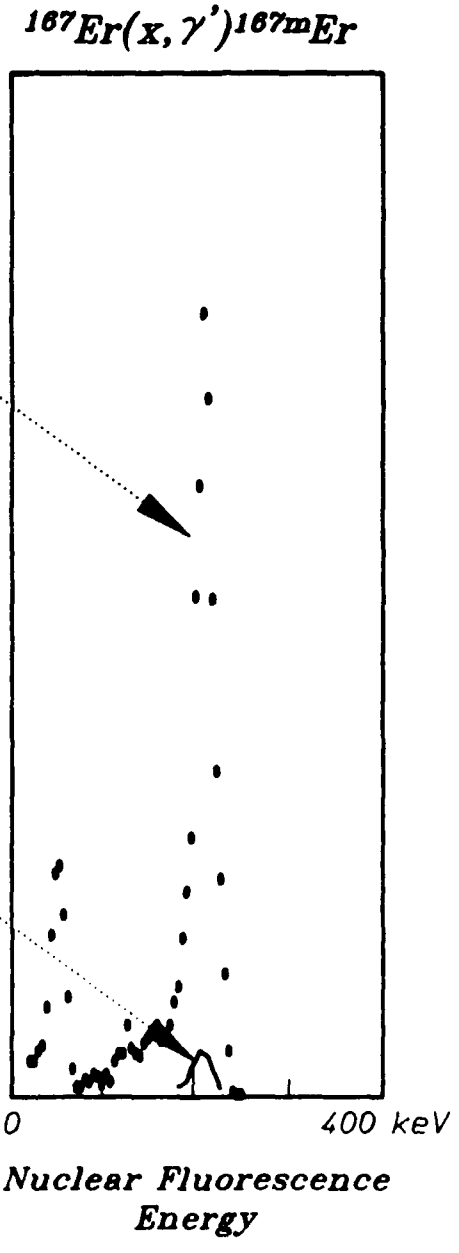
The University of Texas at Dallas
Center for Quantum Electronics
The Gamma-Ray Laser Project
Quarterly Report
April-June 1987



Report GRL/8701

PROOF OF THE FEASIBILITY OF COHERENT AND INCOHERENT SCHEMES FOR PUMPING A GAMMA-RAY LASER

Principal Investigator: Carl B. Collins
The University of Texas at Dallas
Center for Quantum Electronics
P.O. Box 830688
Richardson, Texas 75083-0688

July 1987

Quarterly Technical Progress Report
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Prepared for
INNOVATIVE SCIENCE AND TECHNOLOGY DIRECTORATE
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Contracting Officer's Technical Representative
Dr. Paul Kepple, Code 4720
Naval Research Laboratory
4555 Overlook Avenue, SW
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Recent approaches to the problem of the gamma-ray laser have focused upon upconversion techniques in which metastable nuclei are pumped with long wavelength radiation. At the nuclear level the storage of energy can approach tera-Joules (10^{12} J) per liter for thousands of years. However, any plan to use such a resource for a gamma-ray laser poses problems of a broad interdisciplinary nature requiring the fusion of concepts taken from
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20. Abstract (continued)

relatively unrelated fields of physics. Our research group has described several means through which this energy might be coupled to the radiation fields with cross sections for stimulated emission that could reach 10^{-17} cm^2. Such a stimulated release could lead to output powers as great as 3×10^{21} Watts/liter. Since 1978 we have pursued an approach for the upconversion of longer wavelength radiation incident upon isomeric nuclear populations that can avoid many of the difficulties encountered with traditional concepts of single photon pumping. Recent experiments have confirmed the general feasibility and have indicated that a gamma-ray laser is feasible if the right combination of energy levels and branching ratios exists in some real material. Of the 1886 distinguishable nuclear materials, the present state-of-the-art has been adequate to identify 29 first-class candidates, but further evaluation cannot proceed without remeasurements of nuclear properties with higher precision. A laser-grade database of nuclear properties does not yet exist, but the techniques for constructing one are currently being developed. Resolution of the question of the feasibility of a gamma-ray laser now rests upon the determination of: 1) the identity of the best candidate, 2) the threshold level of laser output, and 3) the upconversion driver for that material.

This quarter's report focuses upon continued development of one of the new technologies being developed for the screening of the laser candidates. It is the nuclear analog of the optical double resonance methods which produced much of the database at the molecular level that was of such essential use in the development of conventional lasers. This technology supported the major milestone demonstration of nuclear fluorescence in the previous quarter, and the successful scaling of this result by another four orders of magnitude is detailed in this quarterly report.

The significance of the results being reported is that they prove our concept of bandwidth funneling over another higher four orders of magnitude and continue to validate our computer codes. The same codes declare that a gamma-ray laser is feasible if the right combination of energy levels occurs in some real material. Discrepancies are found only in the observation that certain detrimental factors expected in some isotopes by theoreticians are not so severe as expected. For example, we report here an excess yield of nuclear fluorescence of 1700% from ^{167}Er, the most elongated nucleus known and the one supposedly most hindered by K-selection rules, and 22,000% excess yield from ^{191}Ir. The simplest implication at this time is that our computer codes for modeling this type of pumping are far better than the basic nuclear data which can be input.

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PREFACE

This being the first report of activities for FY-87, it seems an appropriate time to reflect upon the major achievements of the last year and upon the promising directions to which they point. Both the dressed state (coherent) scheme and the flash x-ray (incoherent) pumping techniques still appear feasible just as we originally proposed in 1982. However, the latter, being simpler, is developing at a more dramatic rate. Since it is the direct nuclear analog of the Group VI laser, proven successful at the atomic level, this is not at all surprising. Rather, it tends to confirm the identity of the critical path most likely to lead to a gamma-ray laser.

Along this path, the efforts of FY-86 culminated in the major milestone demonstration of substantial amounts of nuclear fluorescence pumped by flash x-rays. Proving our concept of bandwidth funneling, we achieved eleven orders of magnitude more output than could have been obtained by pumping the fluorescent level directly. Such results clearly validate our computer codes, the same codes which declare that *a gamma-ray laser is feasible if the right combination of energy levels occurs in some real material*.

So far, FY-87 has seen conceptualization mature to the point where the three best candidates can be nominated on the basis of programmatic merit. Unfortunately, severe difficulties rule against early production of samples of significant size. However, need for these materials continues to build as further successes are realized along the critical path.

The most dramatic achievements in the first quarter of FY-87 have acc·ued from the "scale-up" of nuclear fluorescence when demonstration targets were pumped with x-rays from a nuclear simulator. The first importance of these new results is that the funneled outputs increased linearly with input for another four orders of magnitude, showing no

evidence of detrimental saturation or loss of efficiency. However, another aspect is of comparable significance. Until now our claims that the nuclear database was inadequate to support laser modeling studies could have been perceived as symptoms of insufficient familiarity with the more arcane aspects of disciplines not traditionally our own. The second importance of the scaling studies is that many of the new outputs defied predictive efforts of even the nuclear disciplinarians. *Clearly, our computer codes for modeling the incoherent pumping are far better than the nuclear data which can be input.*

While all concurrent efforts have advanced during the reporting period covered here, the experimental series conducted with the nuclear simulator at Physics International produced so many results that this entire issue is needed for their description. For the execution of that work our UTD team fielded a relatively sophisticated package supporting two parallel shuttle systems capable of transferring in 1.0 sec the irradiated samples to individual NaI detectors, each resolving both decay time and photon energy. In addition we mounted a third system (a high resolution Ge counter) which permitted the concurrent measurement of additional samples, manually detached from the irradiation source and transferred on a time scale consistent with their longer fluorescent lifetimes.

Our systems were up for better than 95% of the four weeks of available machine time, so we were able to examine a total of 32 isotopes. Many were in special targets fabricated from separated isotopes with a book value of over $0.5 million. The combination of high concentration and successful deployment simply buried us in data which will take some time to analyze completely.

A detailed review of the data and analyses to date is found in the following manuscripts. Continuing the preparation of this report as an "in-house" journal, this series presents material to reflect the individual contributions of the teams of research faculty and graduate students involved in these phases of the research. In this regard I wish to thank all our staff for their splendid efforts in supporting the preparation of these manuscripts to a rather demanding timetable.

- C. B. Collins
- Director
- Center for Quantum Electronics

INTRODUCTION

Levels of nuclear excitation which might be efficiently stimulated in a gamma-ray laser are very difficult to pump directly. To have sharply-peaked cross sections for stimulated emission, such levels must have very narrow widths for interaction with the radiation field. This is a fundamental attribute that has led to the facile criticism that "absorption widths in nuclei are too narrow to permit effective pumping with x-rays."

The same comments were voiced in atomic physics before Maiman's great discovery, and it has been useful to pursue this analogy between ruby and gamma-ray lasers. Identification and exploitation of a bandwidth funnel in ruby were the critical keys in the development of the first laser. There was a broad absorption band linked through efficient "kinetics" to the narrow laser level. At the core of one of our proposals[1] for pumping a gamma-ray laser is the use of the analog of this effect at the nuclear level as shown in Fig. 1. A detailed analysis of this option was reviewed in the previous quarterly report,[2] together with the breakthrough report actually demonstrating the great utility of bandwidth funneling at the nuclear level. Yields of gamma-ray fluorescence in ^{77}Se and ^{79}Br were enhanced by eleven orders of magnitude by this effect.[3]

Also shown in Fig. 1 is a further refinement of the incoherent pumping scheme benefiting from upconversion. As has been often discussed[4,5] upconversion as shown in Fig. 1c has many advantages. Most prior reports have emphasized those tending to enhance performance and efficiencies; however, upconversion also makes threshold itself much more accessible. Higher energy isomers need less pump energy to reach the broad collective states that would optimize bandwidth funneling, and the required pump energies can fall in the range where strong x-ray lines may be found to concentrate the spectral intensity.

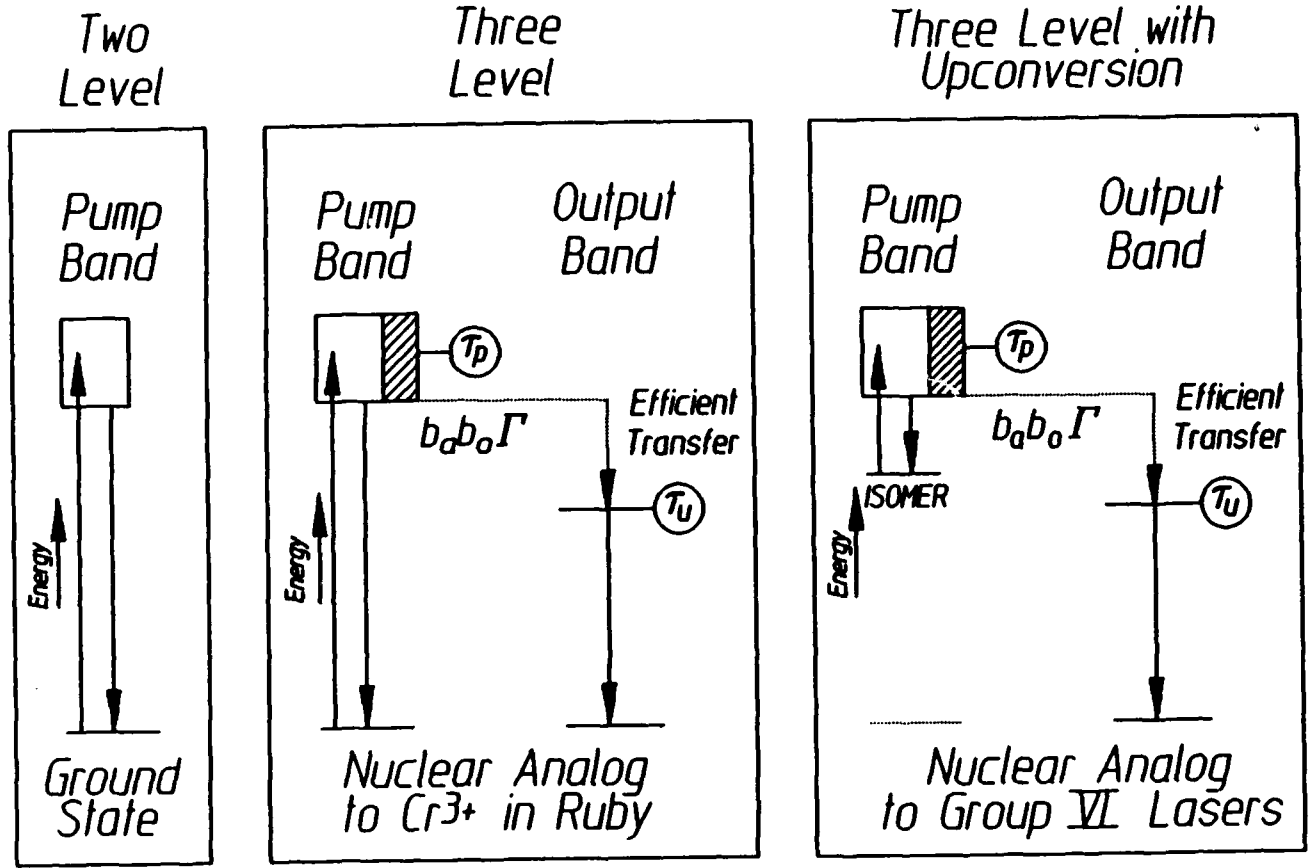

Figure 1: Schematic representation of the energetics of the priority schemes for pumping a gamma-ray laser with flash x-rays. The large width of the level defining the pump band is implied by the height of the rectangle representing the level and the shaded portion indicates that fraction, b_o, which is attributed to the transition to the upper laser level.
(a) Traditional two-level approach.
(b) Three-level analog of the ruby laser serving to illustrate the important concept of bandwidth funneling.
(c) Refinement of the three-level scheme which incorporates upconversion in order to lessen the energy per photon which must be supplied in the pumping step.

In real nuclei, the shortest lifetimes and greatest widths are found for spurious states of collective excitation of nucleons.[6] Instead of a single nucleon generating the transition moment coupling to the radiation field, for these states a collective oscillation of a number of core nucleons contributes a much larger transition moment.

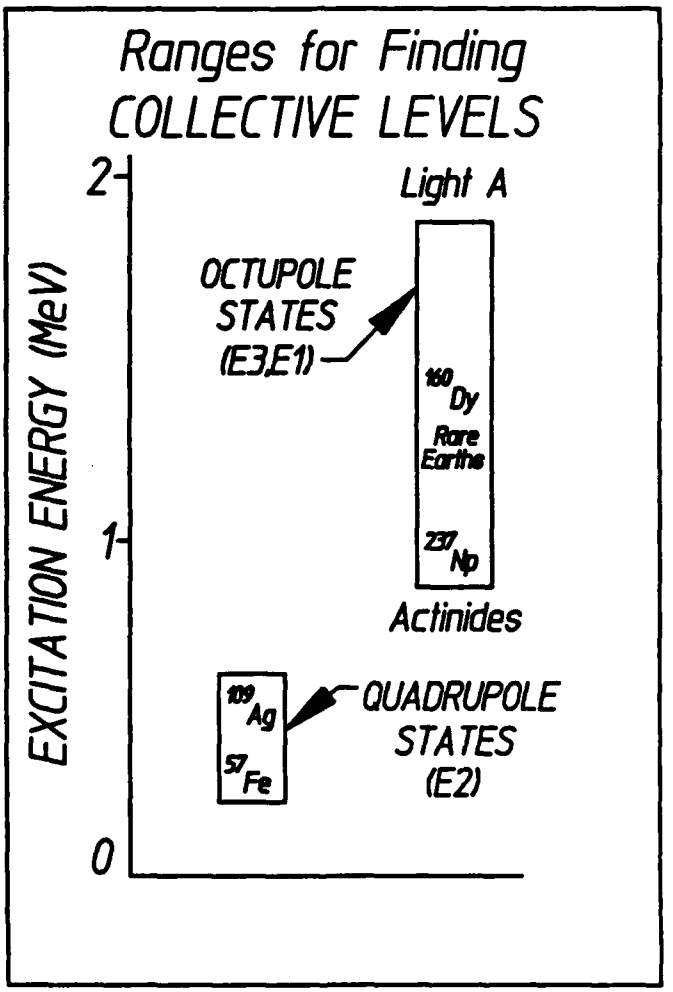

Figure 2: Energy level diagram showing the ranges of excitation above the ground state over which collective oscillations of the nucleonic core may be found. The types of transition moments particularly affected by the oscillations are shown in parentheses.

Over the range of energies of interest to our schemes for pumping a gamma-ray laser, the two types of collective levels shown in Fig. 2

become important. As seen there, the occurrence of collective quadrupole oscillation can be found at the lowest energies. For that case, the important consequence is that the E2 type of pump transition can be considerably enhanced, yielding lifetimes[7] of 1 - 10 psec.

For energies approaching 1 MeV, the possibility of collective octupole oscillation arises. Although octupole, E3 transitions are rarely significant, the symmetry of these oscillations is such that electric dipole, E1 transitions also can be unhindered. This is probably the most important key to the optimization of x-ray pumping schemes. *Collective octupole states near 1 MeV can have lifetimes[8] of 1 - 10 fsec giving effective widths for absorption approaching 1 eV.* This is an enormous width in the context of schemes for pumping a gamma-ray laser.

In evaluating real materials which might serve as the active media in a gamma-ray laser, it is important to be able to relate the fluorescent yield seen in the output transition to the nuclear parameters critical to the schemes of Fig. 1. The general controversy surrounding such computations of yield has been recently resolved[9] both theoretically and experimentally. For a sample which is optically thin at the pump wavelength, the number of nuclei produced in the levels of Figs. 1b and 1c from which the fluorescence will be radiated over the lifetime, τ_u, is given by

$$S = N \sum_i \frac{(\pi b_a b_o \sigma_o \Gamma/2)_i}{E_i} \frac{\phi(E_i)}{A} , \qquad (1)$$

where N is the number of absorbing nuclei in the sample and the summation is taken over the properties and pump fluxes characteristic of each i-th broad pump band centered at pump energy E_i. Only one such pump band is shown in the schemes of Fig. 1, but there could be several at different E_i, each funneling its population into the same output level.

In Eq. (1) the first ratio in the summation is composed of the nuclear parameters, while the second describes the intensities of the pump x-rays which are assumed to be continuous, at least without structure on the fine scale of the nuclear absorption. In particular, the combination $\phi(E_i)/A$ is the spectral fluence at the energy E_i in units of keV/keV/cm^2. Tacitly, it has been assumed[9] that the duration of the pump source is less than the fluorescence lifetime, τ_u.

Of the nuclear parameters, Γ is the natural width in keV of the i-th pump band, as shown in Fig. 1,

$$\Gamma = \hbar/\tau_p \quad , \tag{2}$$

and the branching ratios, b_e and b_o, give the probabilities for the decay of the broad level back into the initial and fluorescence (laser) level, respectively. The pump energy, E_i, is in keV and σ_o is the Breit-Wigner cross section for the absorption transition,

$$\sigma_o = \frac{\lambda^2}{2\pi} \quad \frac{2I_e+1}{2I_g+1} \quad \frac{1}{\alpha_p+1} \quad , \tag{3}$$

where λ is the wavelength in cm of the gamma ray at the resonant energy, E_i; I_e and I_g are the nuclear spins of the excited and ground states, respectively; and α_p is the total internal conversion coefficient for the two-level system shown in Fig. 1a.

Equation (1) is the basic expression for evaluating the number of fluorescent nuclei produced by either of the two pumping schemes of Fig. 1 that can benefit from bandwidth funneling. Were the nuclear parameters accurately tabulated, selection of the best candidate for a gamma-ray laser would be a simple matter of maximizing the terms of Eq. (1). Unfortunately, compilation of the essential parameters is far from adequate, as has been emphasized in previous reports.[2,5,9] Only in very few cases, notably ^{79}Br and ^{77}Se, can the yield be estimated even for the scheme of Fig. 1b. In no case can Eq. (1) be evaluated for the more desirable pumping of Fig. 1c. The only timely recourse is to direct experiments which are modeled on the optical double resonance methods that solved the analogous problems for the conventional lasers at the molecular level.

The ideal experiment would be to pump the candidate nuclei in the upconversion scheme of Fig. 1c and to measure the fluorescence yield directly. The same coupling efficiency from pump to fluorescence is the primary figure-of-merit for evaluating the utility of a candidate in an actual gamma-ray laser. In this way the important combination of variables would be directly measured in such an ideal experiment, and the individual nuclear parameters of Eq. (1) could be determined as byproducts for the subsequent benefit of the nuclear database. Unfortunately again, reality is far from the ideal. The largest sample of any isomer is only 40 mg in mass and contains a concentration of only 4%.

This gives an insufficient value to N in Eq. (1) for a measurable S to be excited with any pulsed x-ray source of laboratory scale. At this time, cost would prevent the preparation of larger samples of any one of the 29 potential candidates.

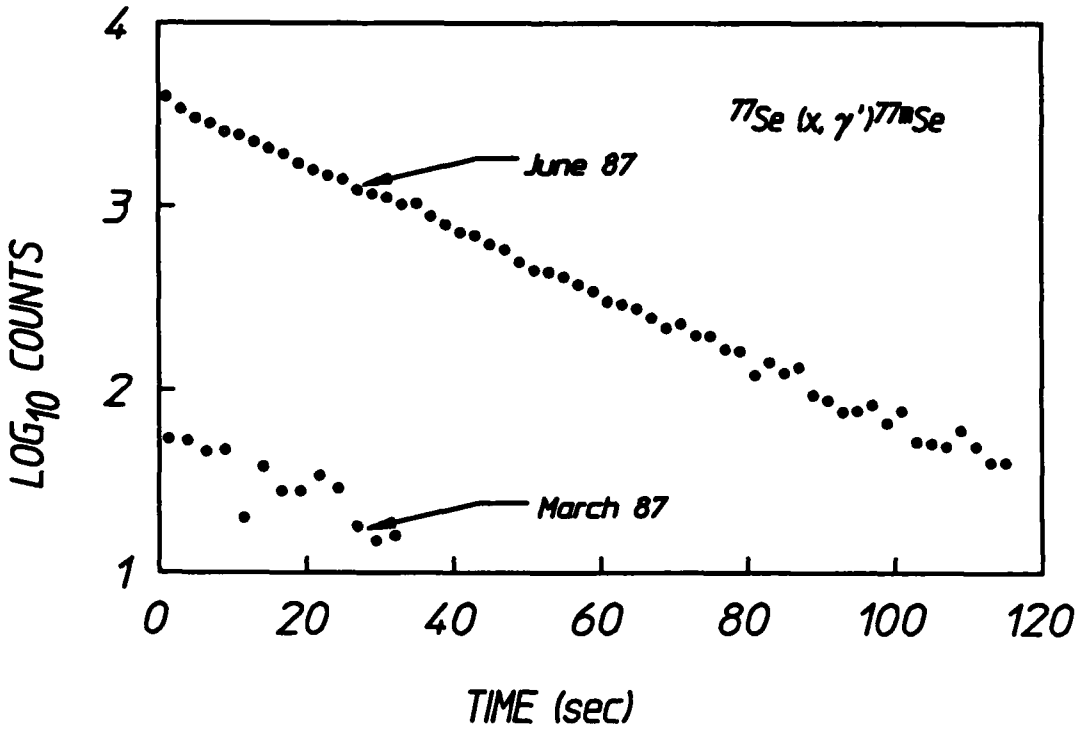

Figure 3: Comparison of the counting rates for nuclear fluorescence from ^{77}Se as functions of time after irradiation for the two experimental series shown; March 1987 pumped by our in-house e-beam machine, APEX, capable of 10^{12} keV/keV and June 1987 pumped by the PITHON nuclear simulator operating around 10^{16} keV/keV. Data from the former were sums taken from 10 discharges into 16.8 g of elemental selenium while the latter were from a single discharge into 3.38 g of isotopically enriched selenium.

A more realistic experiment would be one pumped according to the scheme of Fig. 1b. There, ground state material is used, so sufficient values of N could be reasonably attained. While parameters such as $b_a b_o$ and Γ would not be measured for the laser candidates, themselves, they would be measured for nuclei nearby in the isotopic tables. Subsequent studies of the systematics would certainly improve estimates for the candidates. That, in turn, would greatly facilitate the choice of isomers for production in quantities large enough to evaluate directly.

Such an experiment was done and reported in the previous quarter constituting the major milestone achieved in FY-86.

Described in the following sections is the scaling of this type of fluorescence experiment to much larger pump sources. It comprises the outstanding achievement of the present reporting period. The first importance of these new results is that the funneled outputs increased linearly with input for another four orders of magnitude, as typified by the data reproduced in Fig. 3, showing no evidence of detrimental saturation or loss of efficiency. Shown there is the enhanced output obtained with an order of magnitude fewer shots, and an order of magnitude less material.

A total of 32 isotopes were examined, many in special targets fabricated from separated isotopes. Data were extremely rich in implications. A few examples illustrate this point. The ^{77}Se system is supposed to be completely documented over the appropriate energy range, but we consistently found an excess fluorescence yield of about 40%. More exciting was the excess yield of 1700%, shown in Fig. 4, in ^{167}Er, the most elongated nucleus known and the one predicted to be most hindered by the detrimental K-selection rules. The nucleus ^{197}Au has no known levels connecting to its output channel but fluoresced anyway, as shown in Fig. 5, while ^{189}Os has "known" connections but refused to fluoresce, despite a strong pumping flux at the right energy.

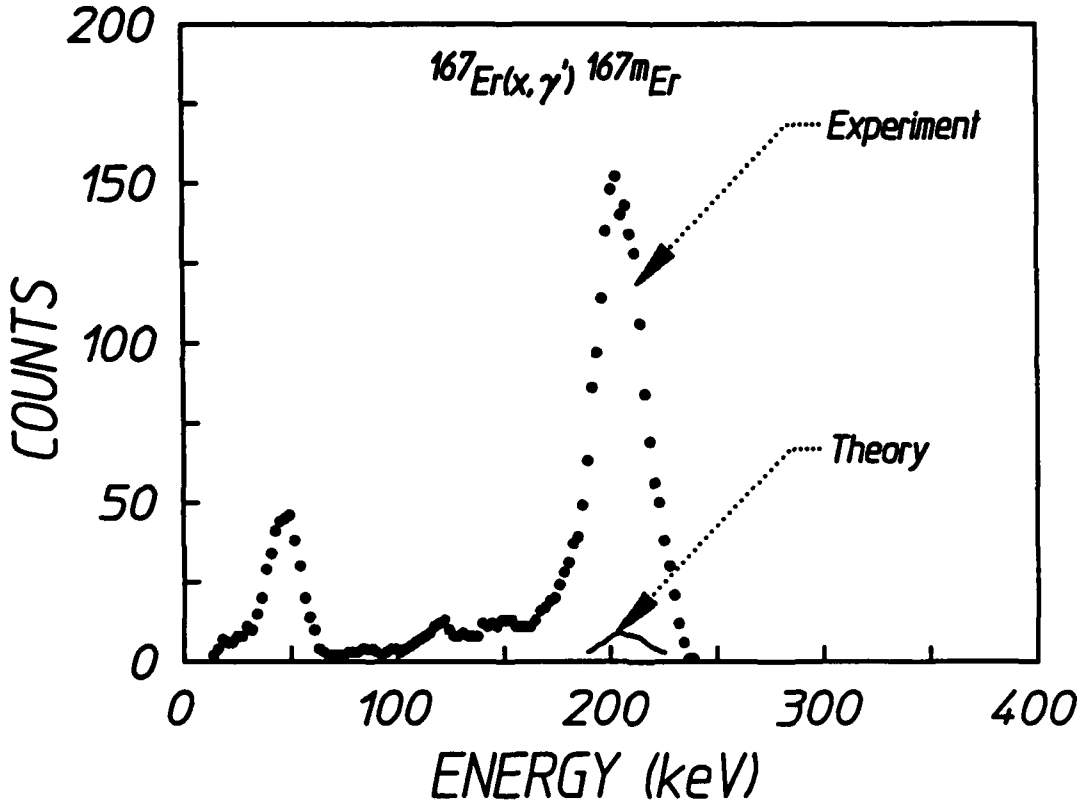

Figure 4: Arrows identify the nuclear component of the spectrum of fluorescence of Er_2O_3 in comparison with model predictions based upon nuclear parameters input from the extant database. Data were obtained from a single irradiation of 2.8 g of Er_2O_3 in natural isotopic abundance.

On each shot the irradiating spectrum was monitored with a standard target of $^{77}Se/^{79}Br/^{115}In$ sampling low, mid, and high pump energies, respectively. Low and mid-energies tended to reproduce from shot-to-shot in accordance with machine specifications for total dose. However, we found that the tail of highest energy x-rays, which contributes only moderately to the dose as determined conventionally, was rather unstable and poorly reproducible. This result seemed to prove the need for this type of selective sampling in order to calibrate the spectra of such pulsed x-ray sources, as recently proposed.[10] In one of the following sections the actual calibration of the pump source is detailed. A subsequent manuscript deals with the peculiar and exciting case of ^{167}Er, together with the many results and implications contributed by the platinum group of isotopes.

A significant conclusion of this quarter's activities is that *our computer codes for modeling the incoherent pumping are far better than the nuclear data which can be input.* This same code has predicted that a gamma-ray laser is definitely feasible if an appropriate isotope exists in reality. This remains the single most critical issue to the development of a gamma-ray laser--the identity of the most nearly ideal candidate for upconversion.

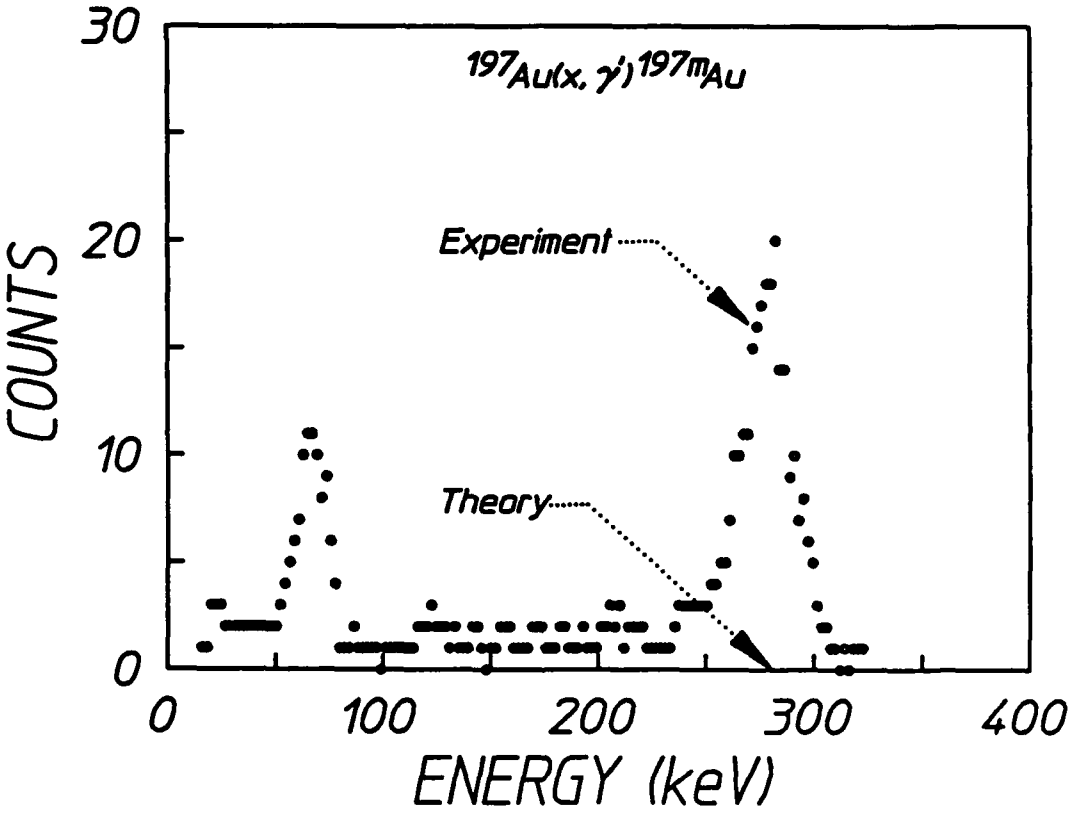

Figure 5: Arrows identify the nuclear component of the spectrum of fluorescence of a gold foil. Zero is expected from models dependent upon input from the extant database of nuclear parameters. Data were obtained from a single irradiation of 2.4 g of elemental gold occurring 100% in the isotope being activated.

References

1. Both the dressed state (coherent) scheme and the flash x-ray (incoherent) pumping techniques still appear feasible just as we originally proposed in C. B. Collins, F. W. Lee, D. M. Shemwell, B. D. DePaola, S. Olariu, and I. I. Popescu, J. Appl. Phys. 53, 4645 (1982). However, the latter, being simpler, is developing at a more dramatic rate.
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CALIBRATION OF PULSED X-RAY SPECTRA

by J. A. Anderson and C. B. Collins

While particle reactions are routinely used for the precise measurement of nuclear parameters, the same levels of accuracy have not been reached in the applications of (γ,γ') reactions. Although examples have been known for over 50 years,[1,2] only a few tens of papers can be found in the literature[3] and results reported for the same reactions show extreme variance. To the nonspecialist this would seem surprising because the analogous optical double resonance techniques are among the most powerful investigative tools at the molecular level.

The principal impediment to the quantitative study of (γ,γ') reactions seems to have arisen from the difficulties in calibrating the sources. Both accelerators and radioactive decay schemes have been traditionally used to provide electromagnetic excitation to reaction sequences such as shown in Fig. 1. Unfortunately, the combination of high flux and high photon energy in the excitation step has not permitted the use of standard spectroscopic techniques, and thus the target has had to be cycled between the irradiation device and the counting facility. In practice this has meant that studies were limited to the excitation of isomers with τ_u lifetimes of seconds to hours.

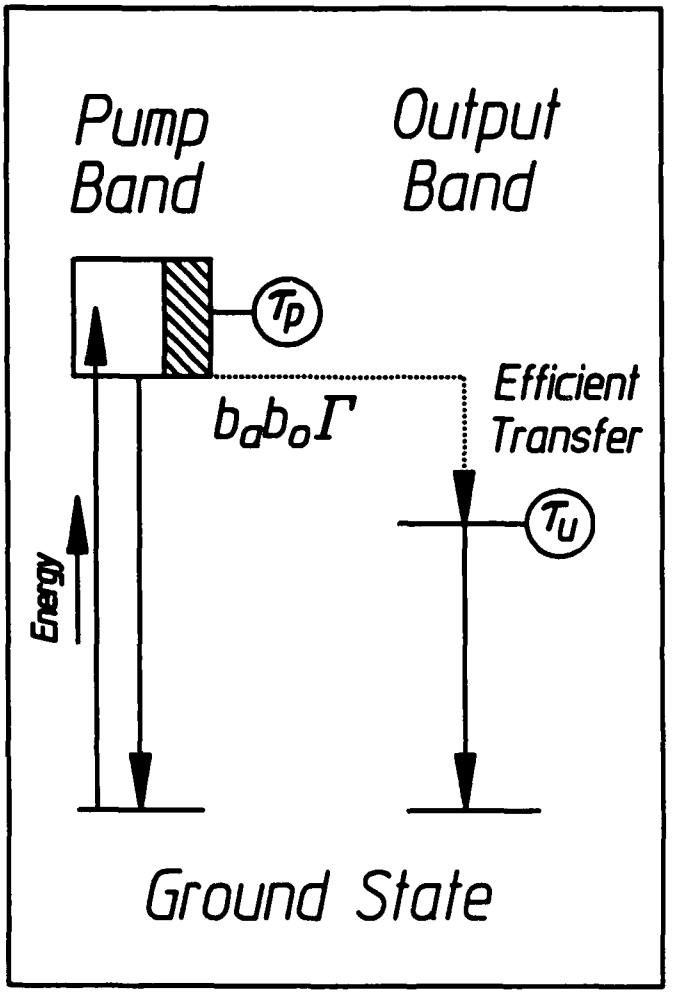


Figure 1: Schematic representation of a typical (γ,γ') reaction sequence which could be used to populate an isomeric state with lifetime τ_u. Absorption of the gamma is assumed first to excite a state with a much shorter lifetime τ_p which then either decays back to the original ground state with probability b_a or to the isomer with probability b_o.

The difficulties in conducting (γ,γ') reactions are probably typified best by the excitation of the 269 minute isomer of ^{115}In at 336 keV. The principal gateway state that is radiatively connected to both the initial and the final isomeric level lies at 1078 keV and can be conveniently excited by resonant absorption of the Compton continuum from a ^{60}Co source mounted in close proximity. Nevertheless, an experiment reported[4] in 1981 produced such an excess of isomeric population that it was necessary to postulate a new mechanism in the ^{115}In for

nonresonant absorption of the undegraded lines from the ^{60}Co source. Theory has not yet provided a process of the needed magnitude, and the most recent repetition of this work[5] has suggested that the excessive yield resulted not from dominance of an unknown process but rather from the difficulty in calibrating the effective dose in the input channel. In that latest, 1986 experiment, excess production was not observed.[5] Such a chaotic level of contradiction between theory and experiment and among the experiments themselves is typical in the literature and attests to the need for a better means for the calibration of intense sources of continuum radiation at energies above 200 keV.

Because of the absence of dispersive materials for such energies and because of the similarity of the absorptive properties contributed by the electrons of the various elements above 200 keV, the absolute calibration of pulsed electromagnetic continua has been virtually impossible. All large-scale sources are currently "calibrated" by fitting a standard theoretical form for the intensity of bremsstrahlung continua as a function of energy to the end point energy and to the total dose.

Unfortunately, dosimeters are not uniform in their response to ionizing radiation. They are much more sensitive to the lower energy photons and thus weight by a large factor the area under the portion of the spectrum most likely to be distorted by effects of self-absorption which, themselves, have a complex dependence upon geometry. The consequence is that such "calibrations" are very sensitive to the particular structure of the spectrum at the low energies and so, in turn, are strongly affected by the accuracy of the parameterization of the individual converter being employed. It is not surprising that discrepancies as great as those affecting the ^{115}In experiments with ^{60}Co sources result from even the most careful efforts with accelerators.

A more direct means of calibration is offered by our approach to the nuclear analog of optical pumping.[6,7,8] There are a few nuclei which are known to have absorption resonances which are broad enough to channel large populations to readily detected states but which remain narrow on the scale of spectral structure of available sources. These few nuclei have been characterized by other types of nuclear studies and can be used to sample narrow slices of the intensity of a continuum. We recently demonstrated[6] the efficacy of this technique by activating targets of ^{79}Br and ^{77}Se for the characterization of a pulsed source of

bremsstrahlung continua at spectral intensities of the order of 10^{12} keV/keV. Excitation was provided by an in-house pulsed electron beam generator having a nominal end point energy of 1 MeV. Accuracy was dependent upon the precision assumed for the tabulated values[9] of nuclear parameters.

Reported here is an extension of this technique to the larger range of photon energies, 0.2 to 1.5 MeV, and for intensities to 10^{16} keV/keV using the same target nuclei, ^{79}Br and ^{77}Se. In this work the self-consistency of the nuclear parameters was directly determined. Important changes and additions were found to be necessary and these are reported. The resulting tabulation of the essential nuclear parameters is now self-consistent and capable of supporting the characterization of pulsed x-ray fluence over the larger range of energies and intensities.

Experimental Method

A schematic drawing of the apparatus used to calibrate the PITHON nuclear simulator at Physics International is shown in Fig. 2. Although only two counting systems are shown for the purposes of illustration, up to three samples could be irradiated during the x-ray pulse and then automatically transferred to the counters. The target-to-counter transit times for the pneumatic shuttles were measured for each shot, and averaged about 1.0 second.

For the work described here, a single sample which incorporated a mixture of LiBr and elemental selenium was used. Figure 3 shows the construction of the sample and lists the important parameters relating to it. After arrival at the counting station, the sample was analyzed for 80 seconds, which represented about four half-lives of the 17.4 second ^{77m}Se and 16 half-lives of the 4.8 second ^{79m}Br. Two NaI(Tl) spectrometers and one HPGe system were employed in the experimental series, but the data presented here was obtained exclusively with one of the 3" x 3" NaI(Tl) detectors. Samples with longer half-lives (e.g. 4.49 hour ^{115m}In) were manually inserted in the spectrometers after the short lived materials were counted.

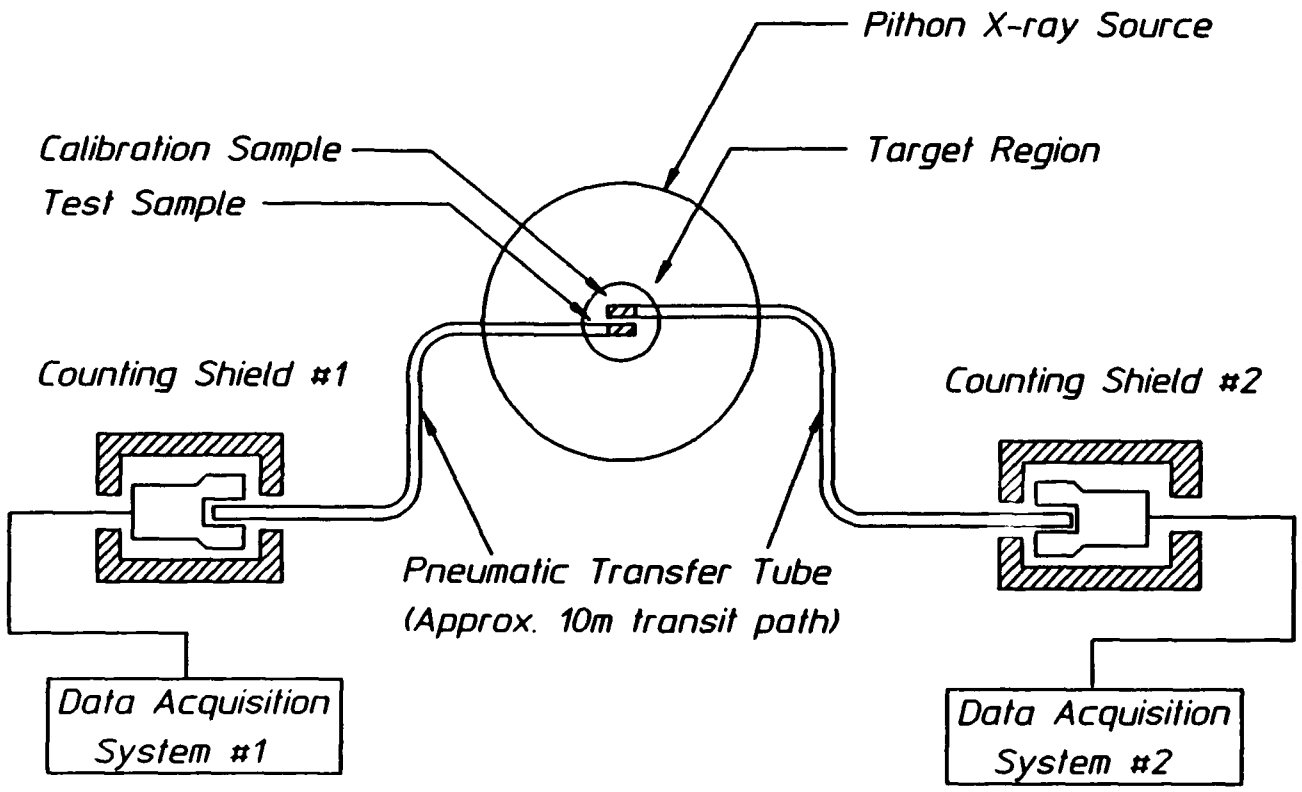
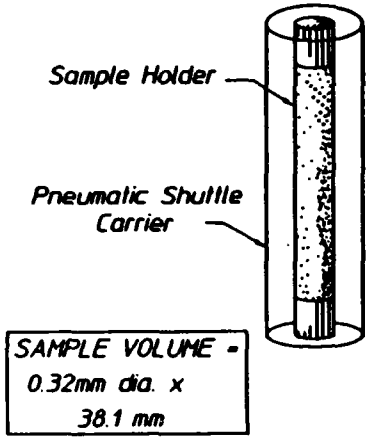


Figure 2: Apparatus for nuclear fluorescence measurements with the PITHON bremsstrahlung machine. Most of the x-ray energy was emitted in a 20 cm pattern in which the samples were centered. The PITHON sequence timer was used to trigger the automatic transfer of samples from the irradiation volume to the counters.



NUCLIDE	FORM	MASS (g)	COUNT TIME (sec)	TRANSIT TIME (sec)
^{79}Br	LiBr	1.250	80.0	1.02 ± 0.03*
^{77}Se	Elemental Se	1.196		

* INDIVIDUAL TRAVEL TIMES WERE USED TO CORRECT EACH TRIAL.

Figure 3: Sample holder for mixed LiBr/Se target. The sample itself was in a 0.64 cm diameter tube capped with plastic plugs. This tube was centered in a polyethylene "rabbit" running in a 1.9 cm diameter transfer tube.

Results and Analyses

During the period of these experiments, 23 shots were successfully instrumented with the calibration target and pneumatic shuttles. The nominal variation of 20% in source performance was augmented by the deliberate programming of firing parameters. As a result, shots were obtained for end point energies ranging from 0.9 to 1.5 MeV. A spectrum of the fluorescence obtained from a typical irradiation is shown in Fig. 4.

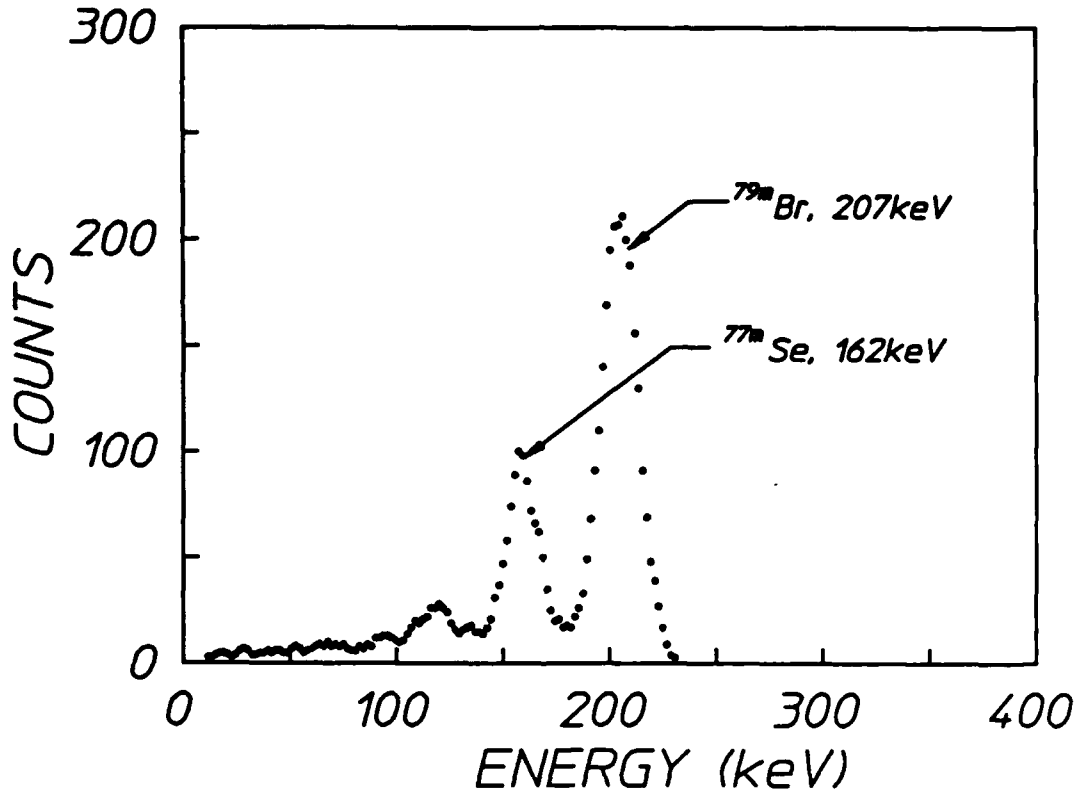


Figure 4: Fluorescence spectrum from a target containing 1.25 g LiBr and 1.20 g of elemental Se, both in natural abundances, excited with a single irradiation (Shot 4381) by the bremsstrahlung produced by the PITHON electron beam device. Acquisition time of this data was 80 sec. Prominent lines are contributed by the isomeric transitions indicated.

Of paramount concern in the examination of data such as shown in Fig. 4 is whether the relative yields from ^{79}Br and ^{77}Se are in agreement with the accepted values for the nuclear parameters appearing in the most recent tabulation.[10] The first step in the resolution of such a question is the extraction of the relative numbers of excited nuclei actually produced in the sample. This requires correction for the finite duration of the sampling period over which the fluorescence is counted, the efficiency of the detector, the probabilities for internal conversion rather than radiation of the fluorescence, and the fraction of fluorescence reabsorbed in the sample. While the first three corrections are readily obtained from tables or by calibration, the last is dependent upon geometry in a more complex fashion. To minimize the importance of that particular correction, small samples were used for which the effects on the ^{79}Br and ^{77}Se data differed[11] by about 3%.

As described in the article[6] reporting the previous implementation of this technique at lower energies, the number of excited nuclei produced in the course of an irradiation of the type shown in Fig. 1 can be computed from the expression:

$$S = N \sum_i \frac{(\pi b_e b_o \sigma_o \Gamma/2)_i}{E_i} \quad \frac{\phi(E_i)}{A} \quad , \qquad (1)$$

where N is the number of absorbing nuclei in the sample and the summation is taken over the properties and pump fluxes characteristic of each of the i-th broad pump bands centered at pump energy E_i. Only one such pump band is shown in the scheme of Fig. 1, but there could be several at different E_i, each funneling its population into the same output level.

In Eq. (1) the first ratio in the summation is composed of the nuclear parameters, while the second describes the intensities of the pump x-rays which are assumed to be continuous, at least without structure on the fine scale of the nuclear absorption. In particular, the combination $\phi(E_i)/A$ is the spectral fluence at the energy E_i in units of keV/keV/cm^2. Tacitly, it has been assumed that the duration of the pump source is less than the fluorescence lifetime, τ_u.

Of the nuclear parameters, Γ is the natural width in keV of the i-th pump band, as shown in Fig. 1,

$$\Gamma = \hbar/\tau_p \quad , \qquad (2)$$

and the branching ratios, b_e and b_o, give the probabilities for the decay of the broad level back into the initial and fluorescence level, respectively. The pump energy, E_i, is in keV and σ_o is the Breit-Wigner cross section for the absorption transition,

$$\sigma_o = \frac{\lambda^2}{2\pi} \frac{2I_e+1}{2I_g+1} \frac{1}{\alpha_p+1} , \qquad (3)$$

where λ is the wavelength in cm of the gamma ray at the resonant energy, E_i; I_e and I_g are the nuclear spins of the excited and ground states, respectively; and α_p is the total internal conversion coefficient for the broad level of Fig. 1.

The nucleus ^{79}Br is unique in its utility to efforts at calibration. According to latest data,[10] it has but a single broad level connecting to both ground and fluorescent states in the energy range 0 - 1.8 MeV. The relevant part of its scheme of energy levels is shown in Fig. 5. On the other hand, the ^{77}Se nucleus is more complex having at least four levels funneling to the same fluorescent transition at 161.8 keV, as shown in Fig. 5. The highest at 818 keV is insufficiently characterized, and the Γ_i parameter appearing in Eq. (1) must be estimated. This was done and reported previously.[6,7] The "best" values[12] of the parameters of Eq. (1) are listed in Table I for convenience, together with sources of the basic data and the values confirmed by this experiment.

An additional difficulty encountered in the use of Eq. (1) to model the performance of targets pumped with the device used in these experiments accrues from the geometry of the diode producing the bremsstrahlung x-rays. Since it is configured as a pinched diode, electrons do not arrive at the converter foil with normal incidence. In such cases the linear, Kulenkampff approximation to the spectrum used previously[6] is expected to become markedly concave.[13]

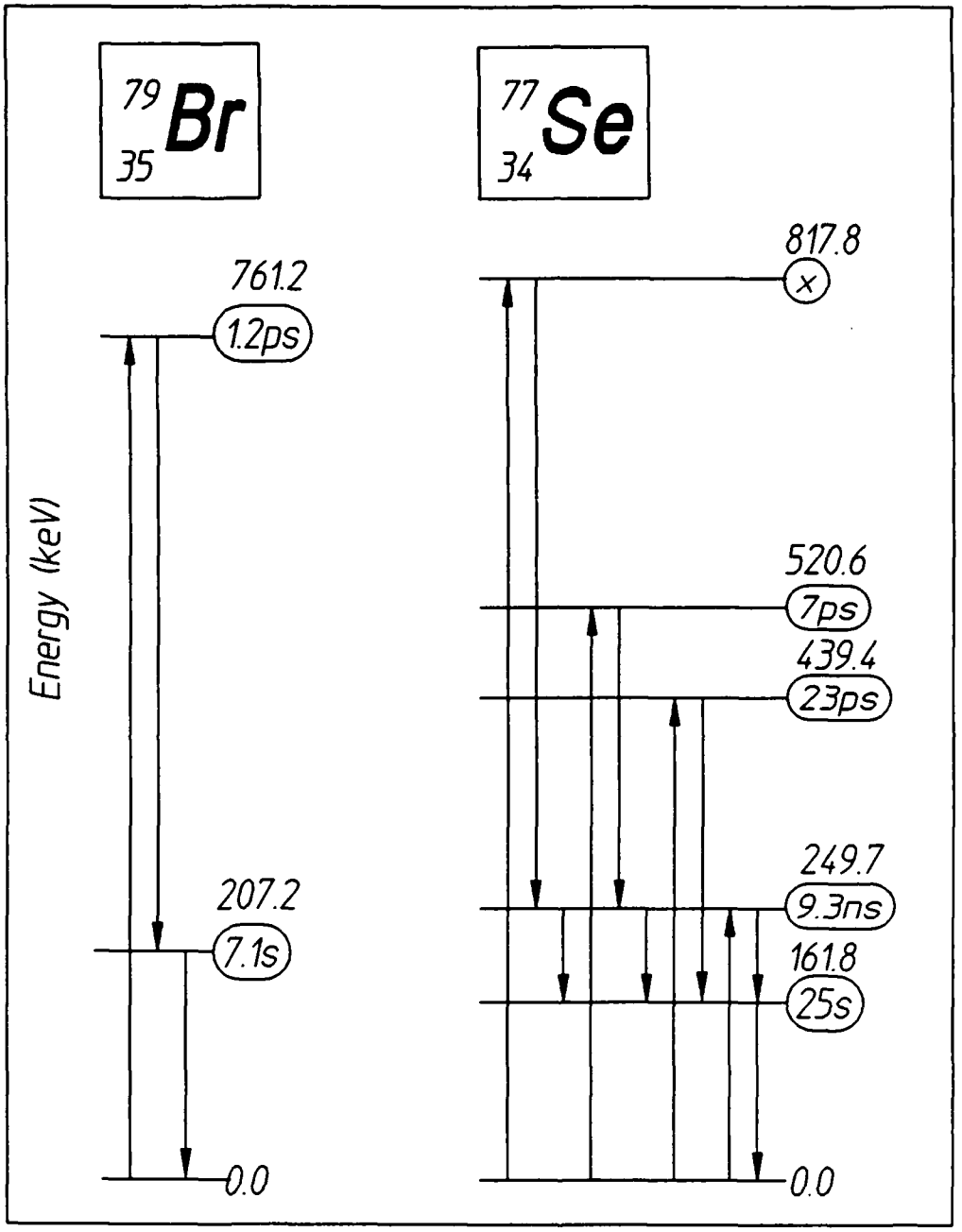


Figure 5: Energy level diagram of the excited states of ^{79}Br and ^{77}Se important in the production of populations of their respective isomers. Half-lives of the states are shown in the ovals to the right of each and sequences of transitions corresponding to the (γ,γ') reactions leading to the isomers are shown by the arrows. Downward γ' transitions also include effects of cascading through levels other than those shown.

Table I

Summary of nuclear fluorescence parameters reconciled by this work.

NUCLIDE	E_{in} (keV)	$\pi b_a b_o \Gamma \sigma_o/2$ (10^{-29} cm^2 keV)		E_{out} (keV)
		Ref. 6	This work	
^{79}Br	761	6.2	6.2	207
^{77}Se	250	0.20	0.20	162
	480[a]	1.50	0.87	
	818	0.7	0.7	
	1005		30	
^{115}In	1078		20[b]	337

[a]The effects of the 440 keV and 521 keV transitions have been combined.
[b]From Ref. 17.

Of the 23 shots instrumented in this experimental series, five benefited from additional diagnostics. From records of the time dependent voltage and current, the spectral fluence was calculated with the one-dimensional coupled electron/photon Monte Carlo transport code, TIGER, an established procedure in the e-beam community.[14] Results supplied by Physics International are shown in Fig. 6 for a selection of those five shots. If plotted on a linear scale, those spectra would be considerably more concave than the idealized approximation[6] used for normal incidence. However, the ratio of fluence at a particular energy to that at some standard can be readily parameterized from curves such as shown in Fig. 6. In doing this, the energy of the single absorption line of ^{79}Br at 761 keV was chosen as a standard. Shown in Fig. 7 are the resulting curves for the empirical estimation of the relative spectral intensity, $\zeta(E_i)$,

$$\zeta(E_i) = \phi(E_i)/\phi(761 \text{ keV}) \quad , \qquad (4)$$

plotted as functions of the end point energy, V_o, for four different E_i, 250, 480, 818, and 1005 keV. Data points record the values supplied by the TIGER code, and the curves were obtained by a smoothing procedure.

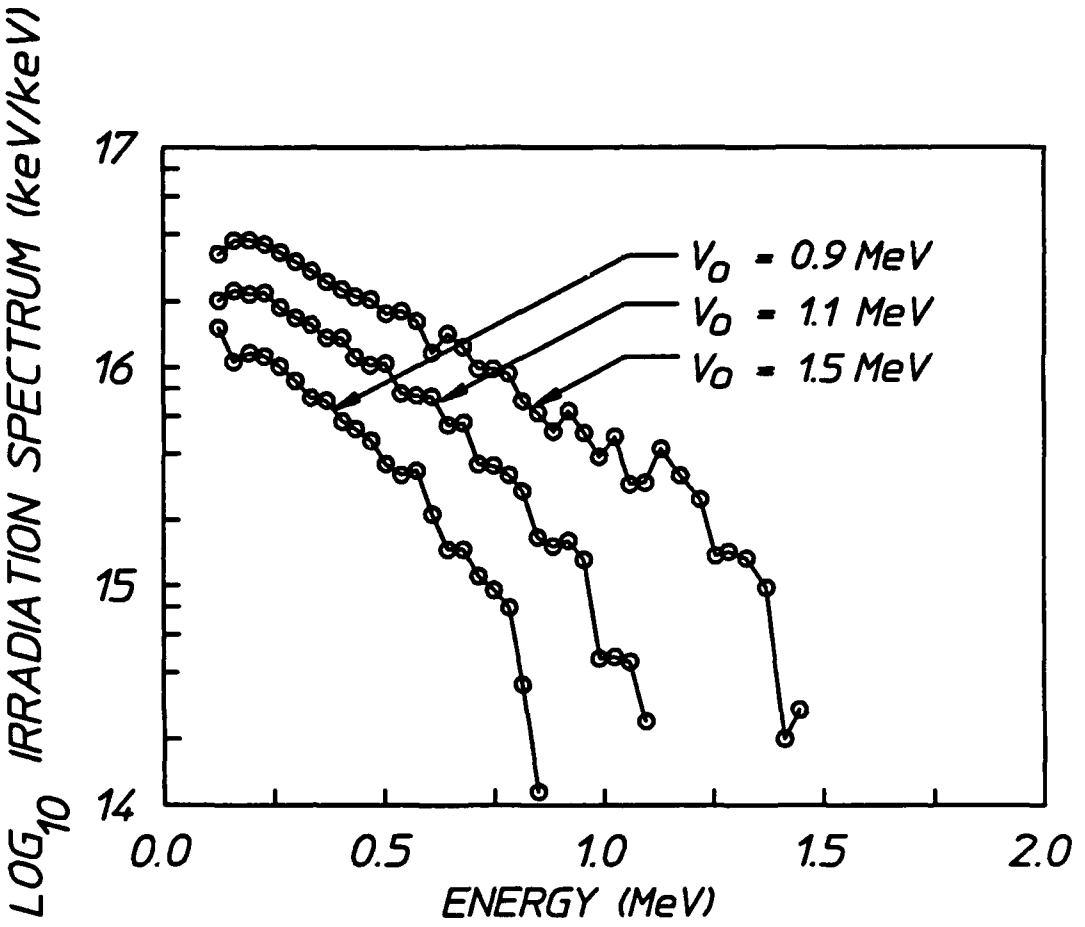


Figure 6: Plot of three bremsstrahlung spectra computed with the TIGER code for the particular characteristics of three electron beam discharges from PITHON having the end point energies, V_o, as indicated.

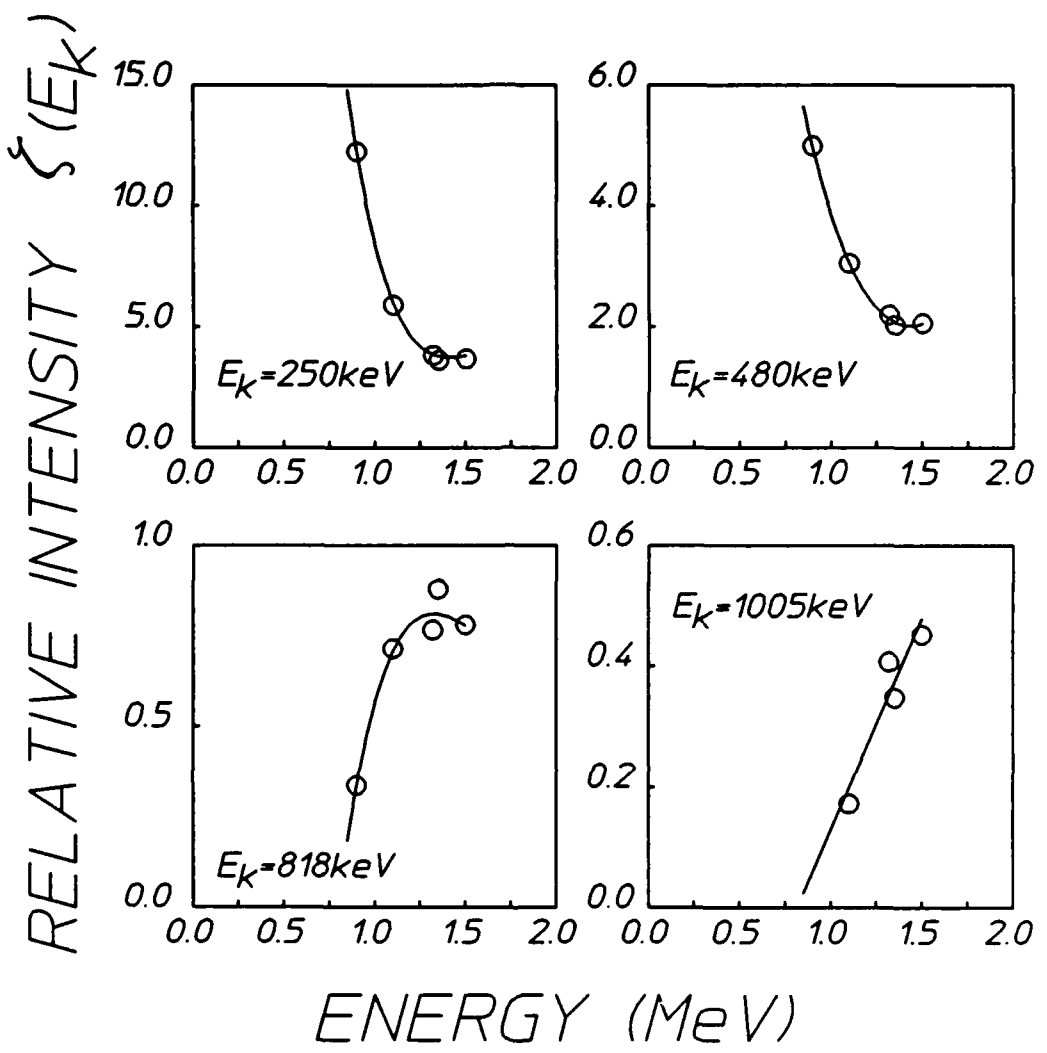


Figure 7: Graphs of the relative spectral intensities $\zeta(E_i)$ at the photon energy, E_i, plotted as functions of the end point energy of the electron beam producing the bremsstrahlung. Curves were obtained by smoothing and interpolating between the data points representing the results of TIGER code computations, three of which are shown in Fig. 6. Intensities are normalized to the spectral intensity at the reference energy E = 761 keV.

From data such as presented in Fig. 7 the fluorescent yield expected from ^{77}Se can be readily computed in terms of that actually observed from ^{79}Br. The degree to which the calculated yield agrees with the measured fluorescence is a direct indication of the level of consistency between the values compiled for the nuclear parameters in Table I. This can be appreciated by writing Eq. (1) for the irradiation of a sample of ^{79}Br,

$$S(Br) = N(Br)\xi_{761}(Br)\frac{\phi(761)}{A} \quad , \tag{5}$$

where

$$\xi_i = \frac{(\pi b_a b_o \sigma_o \Gamma/2)_i}{E_i} \quad , \tag{6}$$

from Eq. (1), and $\phi(761)$ is the spectral intensity *at the target*. The corresponding expression for the ^{77}Se is,

$$S(Se) = N(Se)\left[\xi_{250}(Se)\frac{\phi(250)}{A} + \xi_{480}(Se)\frac{\phi(480)}{A} + \xi_{818}(Se)\frac{\phi(818)}{A} + \xi_k(Se)\frac{\phi(E_k)}{A}\right] \quad , \tag{7}$$

where the possibility of an extra contribution from an unexpected band at E_k has been included.

Dividing Eq. (7) by Eq. (5) and substituting from Eq. (4) and (6) yields after some rearrangement,

$$\frac{S(Se)}{S(Br)} - \frac{N(Se)}{N(Br)}\left[\frac{\xi_{250}(Se)}{\xi_{761}(Br)}\zeta(250) + \frac{\xi_{480}(Se)}{\xi_{761}(Br)}\zeta(480) + \frac{\xi_{818}(Se)}{\xi_{761}(Br)}\zeta(818)\right] = \frac{N(Se)}{N(Br)}\frac{\xi_k(Se)}{\xi_{761}(Br)}\zeta(E_k) \quad . \tag{8}$$

While formidable in appearance, Eq. (8) has a very straightforward interpretation, namely,

$$R(\text{exp}) - R(\text{model}) = \frac{N(Se)}{N(Br)} \frac{\xi_k(Se)}{\xi_{761}(Br)} \zeta(E_k) \quad , \qquad (9)$$

where R is the ratio of the numbers of excited nuclei produced in ^{77}Se and ^{79}Br.

Several points warrant comment. While $\phi(E_i)$ in Eq. (5) depends upon the poorly-characterized geometric efficiency through which the fluences of Fig. 6 are actually coupled to the target, the relative fluence, $\zeta(E_i)$ does not. Neither does it depend upon the entrance aperture of the target, A, another uncertain quantity in real configurations.

In actual experiments, the residues between experimental data and the results of the model appearing on the left of Eq. (9) should be scattered about zero, regardless of the end point energy of the irradiation--*provided the model is complete*. If an unknown channel is contributing, or if one of the $\xi_i(Se)$ is incorrect, then the residue should have a dependence upon experimental variables as shown to the right of Eq. (9). Since the $\zeta(E_k)$ of Fig. 7 have considerably different functional dependences upon V_o, it should be possible to identify the particular E_k contributing the residue, provided enough variation of V_o can be introduced into the experiment.

For the 23 data points of this experiment, the residues computed from Eq. (9) do <u>not</u> scatter about zero. Figure 8 shows a plot of the resulting residues as functions of end point energies. The functional dependence is striking and fits a line intercepting the horizontal axis near 1 MeV. Inspection of the right side of Eq. (9) suggests that this would be consistent with the contribution from an additional transition with a threshold energy for excitation near 1 MeV.

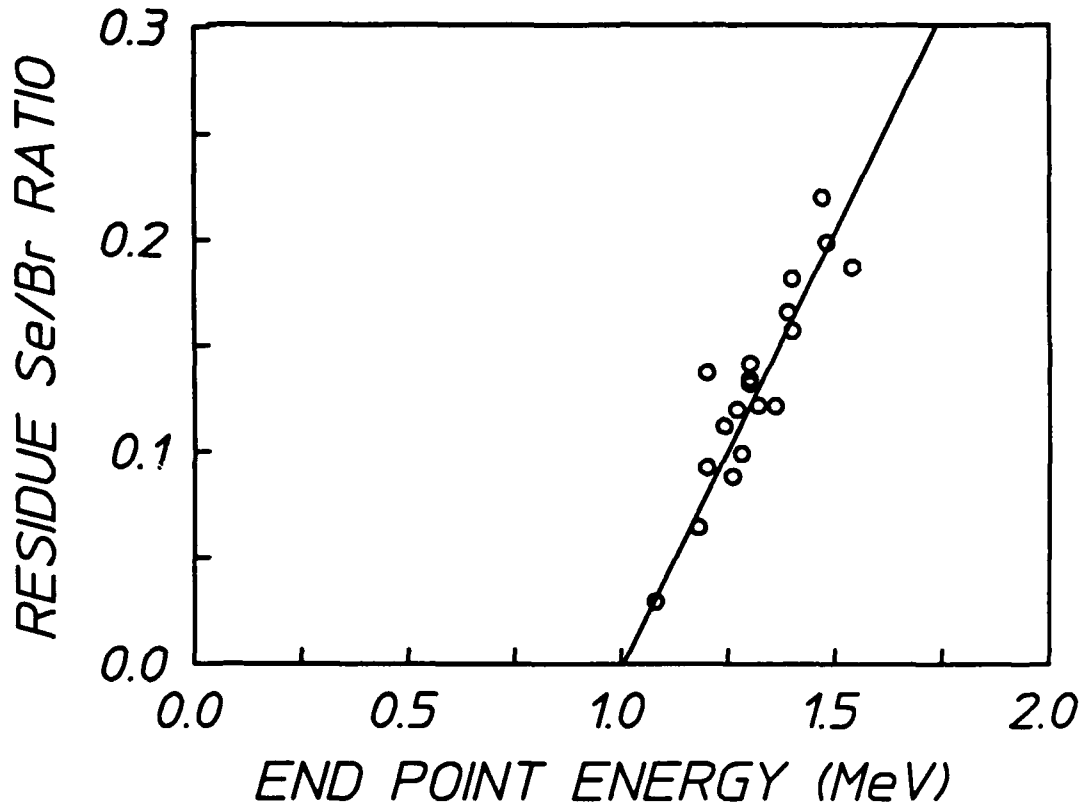


Figure 8: Plot of the residues obtained from the application of Eq. (9) to the data for a model containing the three lowest energy gateway levels of ^{77}Se plotted as a function of the end point energies of the electron beam discharges producing the bremsstrahlung.

The complete set of nuclear parameters appearing in Eq. (6) is not known for any of the transitions of ^{77}Se lying above 818 keV. However, the branching ratios are available for many of them. While optimal branching ratios could be offset by poor lifetimes, in principle, it is most reasonable to expect a significant new channel of excitation to correspond to a level for which branching ratios were, at least, favorable. Table II records the product $b_a b_o$ for levels of ^{77}Se above 818 keV, and the close correlation between the large value seen at 1005 keV and the intercept of Fig. 8 is extremely persuasive.

Table II

Compilation from Ref. 10 of the products of the branching ratios, $b_a b_o$, appearing in Eq. (6) for the transitions of ^{77}Se lying above 818 keV. Corresponding values of Γ have not been reported in the literature.

Level (keV)	$b_a b_o$
818	0.0063
825	0.00025
911	0.017
1005	0.031
1128	0.0012
1186	0.047
1230	0.0024

Once the excitation energy of the "new" channel was determined from Fig. 8 and Table II, the functional dependence of $\zeta(1005)$ upon end point energy could be determined. This was done, and the result is shown in Fig. 7. Again there is a striking similarity between the functional dependences upon V_o of the residues and of the $\zeta(1005)$, as would be required by Eq. (9).

Once the level missing from the model is identified as corresponding to E_k = 1005 keV, all of the terms of Eq. (9) are known except $\xi_{1005}(Se)$. In Fig. 9 the model residues are plotted as functions of $\zeta(1005)$ for the 19 shots of this experiment having end point energies above 1 MeV. From Eq. (9) it can be seen that the slope of such a plot should correspond[15] to $\xi_{1005}(Se)N(Se)/\xi_{761}(Br)N(Br)$. The least-squares fit to the data including the origin is shown by the middle diagonal line in Fig. 9, together with the outer lines bounding acceptable alternatives. These lead to a value

$$\xi_{1005}(Se) = (30 \pm 7.5) \times 10^{-32} \text{ cm}^2 \quad , \qquad (10a)$$

which from Eq. (6) gives,

$$\pi b_a b_o \sigma_o \Gamma/2 = (30 \pm 7.5) \times 10^{-29} \text{ cm}^2 \text{ keV} \quad , \qquad (10b)$$

in turn yielding,

$$t_{½}(1005) = 0.36 \text{ psec} \quad . \qquad (10c)$$

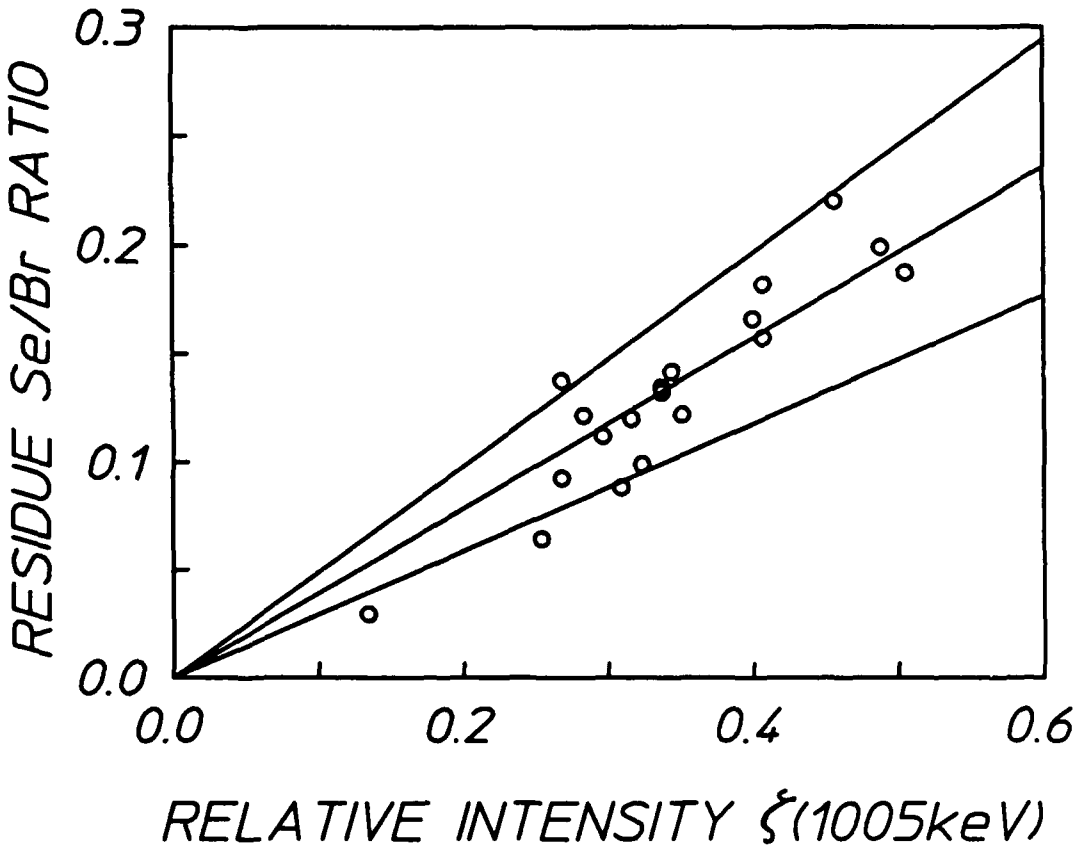


Figure 9: Plot of the residues of Fig. 8 as functions of the relative intensity of irradiation at 1005 keV.

As a final step of analysis, the residues of Eq. (9) were recomputed, including the term of Eq. (10a) into the model estimate, R(model), on the left of Eq. (9). Figure 10 shows the resulting residues as functions of the end point energies. This time the data appear[16] to scatter around the V_o axis indicating that the model now contains a sufficient number of terms to predict the fluorescent yields up to an end point energy of 1.5 MeV to an accuracy of better than 10%.

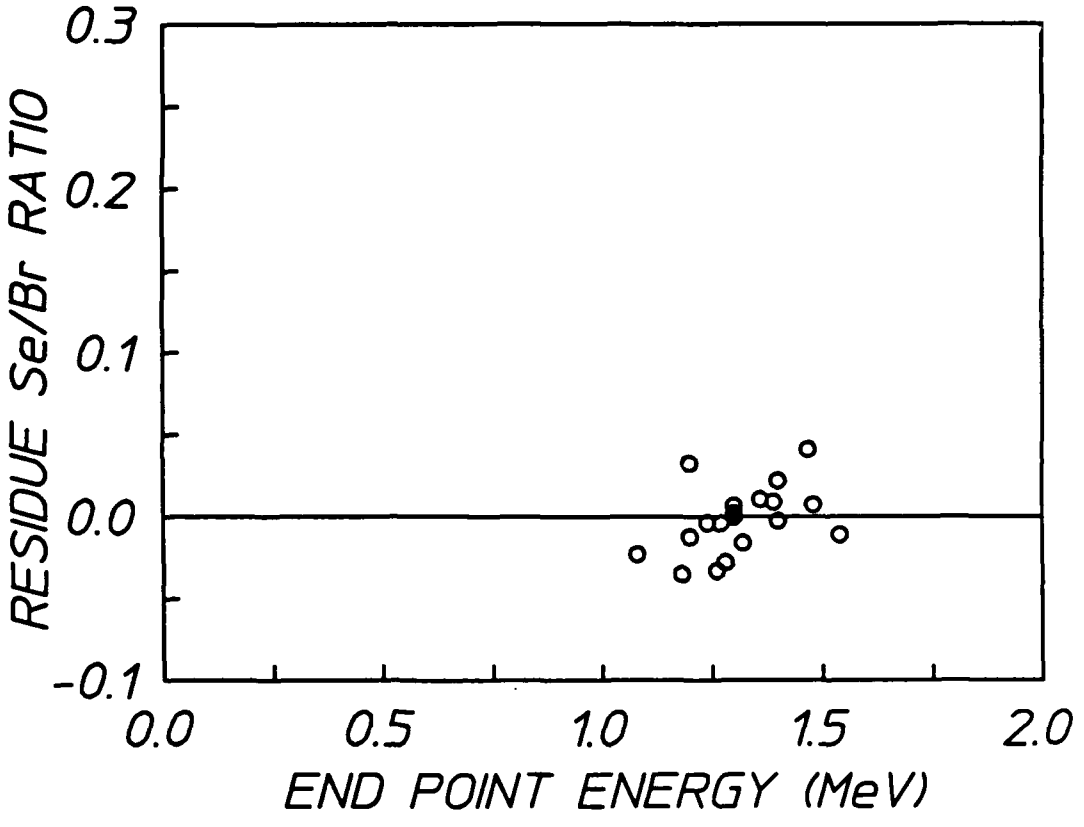


Figure 10: Plot of the residues obtained from the application of Eq. (9) to the data for a model containing the three lowest energy gateway levels of ^{77}Se together with the new level at 1005 keV, plotted as a function of the end point energies of the electron beam discharges producing the bremsstrahlung.

Discussion

The principal conclusion of this work is that the 1986 version[10] of the nuclear parameters for ^{77}Se and ^{79}Br is completely consistent. Provided no significant component of the x-ray flux lies above 1 MeV, those data are sufficient to predict the fluorescent yields from ^{79}Br and ^{77}Se. However, for the irradiation of samples with bremsstrahlung produced by electrons with end point energies above 1 MeV, the more complete set of parameters of Table I must be used. In such cases it is appropriate for all end point energies below 1.5 MeV.

The difference between the 1986 data and Table I lies in the inclusion of the pump channel in ^{77}Se at 1005 keV found in the course of this work. Corresponding to the considerable transition strength of 0.011 Weisskopf units for an M1 transition in the absorption channel, it is well-connected to both initial and fluorescent levels. As can be seen from Table I it will rapidly become the dominant channel for funneling population into the fluorescence level as the number of pump photons above 1 MeV increases in a source.

The striking consistency between the results from ^{79}Br and those from ^{77}Se argues favorably for the utility of this technique of selective nuclear excitation over a wider field of experimental variables than originally described.[6] It does not require an initial computer model. A measurement of the activation of ^{79}Br would determine the fluence at 761 keV at the position of the sample. Activation of ^{77}Se is more complex but tractable, once the ^{79}Br results are known. Assuming $\zeta(818) \sim \zeta(761)$ the contribution to ^{77}Se from the small 818 channel can be easily removed. The immediate residue is a weighted sample of the low energy portion of the irradiating spectrum and of the energies near 1 MeV. To separate these effects in cases where the end point energy lies above 1 MeV, recourse must be made to a third nuclei sampling only high energies. An ideal candidate is ^{115}In which is the subject of a forthcoming article.[17] It samples only the fluence at 1078 keV and the consistent value[17] for the corresponding reaction has also been included in Table I for convenience. From the activation of an ^{115}In component of the sample, the contribution to the ^{77m}Se population from the new 1005 keV channel can be removed. From the resulting second residue, the average fluence in the range 250 - 480 keV can be isolated.

As a demonstration of the efficacy of nuclear activation, such a procedure was performed on the activation of a composite target irradiated with a typical shot (4379) from PITHON. As expected, absolute measurements of the flux at the target could be obtained for three energies,[18] 433, 761 and 1078 keV. Then by multiplying those values of flux by the area of the forward (2π) hemisphere upon which the target was conceived to rest during irradiation, the absolute spectral intensity emitted by the source was determined. Results are plotted in Fig. 11 together with the predictions of the TIGER code calculations for that shot. Agreement is perhaps better than is warranted by the inherent level of mechanical inaccuracy in target positioning.

Uncertainty in the direct measurement of spectral intensity at the target arises from two sources, statistical error in the number of fluorescent counts and unknown error in the nuclear data of Table I. Results of the former are plotted in Fig. 11, but appear larger than the plotted symbol only at the lower energy point arising as a result of so many differences of data. Uncertainty due to residual inconsistency in the data of Table I is of unknown magnitude but should be less than the error introduced by the statistical uncertainty in the counts as a result of these efforts in determining consistency of the entries of Table I.

The results of this work indicate that (γ,γ') reactions can be studied with a high level of precision if the sources are carefully characterized. The existing data for the transitions of ^{79}Br and ^{77}Se, together with others to be discussed, clearly offer a convenient means of sampling the spectra of intense pulsed sources.

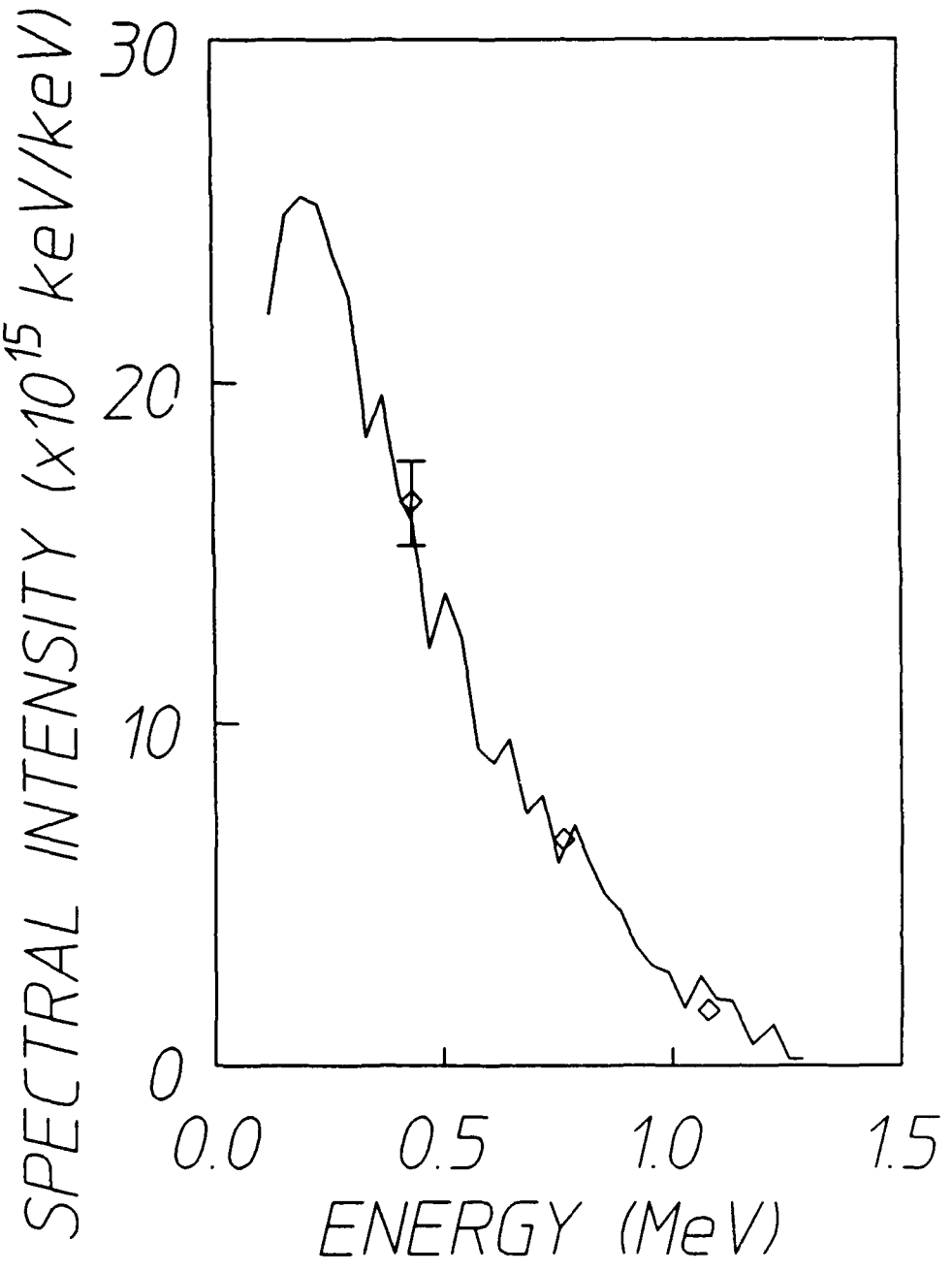


Figure 11: Data points plot the spectral intensities measured directly with the nuclear activation technique using the parameters of Table I in comparison to the spectrum computed with the TIGER code for a typical PITHON shot, 4379. Vertical bars show uncertainty in the measurement at the one point for which that uncertainty was larger than the plotted size of the symbol.

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7. J. A. Anderson and C. B. Collins, Proof of the Feasibility of Coherent and Incoherent Schemes for Pumping a Gamma-Ray Laser, University of Texas at Dallas, Report #GRL/8602, Innovative Science and Technology Directorate of Strategic Defense Initiative Organization, April 1987, pp. 29-46.

8. We recently learned that apparently the first demonstration of the feasibility of the principle of this technique was made in 1971 before tabulations of nuclear parameters had matured to the point of being able to provide adequate practical guidance. See: L. Cohen and E. A. Wolicki, NRL Report 7306 (1971).

9. C. M. Lederer and V. S. Shirley, ed. Table of Isotopes (John Wiley, New York, 1978).

10. Evaluated Nuclear Structure Data File (Brookhaven National Laboratory, Upton, New York, 1986).

11. The actual values for the fractions of the fluorescent photons unaffected by self-absorption were calculated to be 91 and 88% for ^{79}Br and ^{77}Se, respectively.

12. As seen in Table I, the best values of nuclear parameters differ from those used in Refs. 6 and 7. Our earlier reports used data from Ref. 9, while more recently we used Ref. 10, current to 1986.

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14. J. A. Halbleib and T. W. L. Sanford, Sandia Report SAND83-2572 (1983).

15. Data actually plotted in Figs. 8 - 10 are ratios of the numbers of detected photons corrected for the finite period of counting, while the analyses of Eqs. (1) - (9) are written in terms of the ratios of the numbers of active nuclei produced in the target. Slopes obtained from the figures must be multiplied by 1.50, the product of the ratios of corrections for detector and fluorescence efficiencies and for self-absorption in the target before being used in the equations.

16. The scatter of the residues in Fig. 10 does not appear completely random and a trend might be perceived as indicating another gateway opening around 1.2 MeV. It is tempting to identify this with the level at 1186 keV favored in Table II. However, more shots with higher end point energies would be needed to obtain a meaningful assessment of such a possibility.

17. C. B. Collins, J. A. Anderson, Y. Paiss, C. D. Eberhard, J. Peterson, and W. Hodge, Phys. Rev. C. (pending).

18. The first, 433 keV, is the average of the energies of the lowest two transitions in ^{77}Se weighted by their respective ξ_i from Eq. (6). Pragmatically this corresponds to an average line at 433 keV having $\xi_{433}(\mathrm{Se}) = 3.9 \times 10^{-32}$.

OPPORTUNITIES FOR NUCLEAR ACTIVATION

by C. B. Collins and J. A. Anderson

Recent manuscripts[1,2] have reported how effectively the technique of x-ray activation of nuclei (XAN) can be used to calibrate the spectral intensities found in a single pulse of intense bremsstrahlung. Essential in determining the pump intensities used in schemes for exciting a gamma-ray laser,[3] XAN has also been validated as a means of calibrating nuclear simulators.

As currently implemented, three isotopes, ^{77}Se, ^{79}Br, and ^{115}In, are used to sample narrow spectral slices of the fluence illuminating a target. Information is stored as isomeric excitations to be "read out" later. With three isotopes, XAN accommodates measurements at three photon energies, 433, 761 and 1078 keV.

The key to the development of that technique was the use of a large number of shots to activate test samples in order to resolve experimentally the level of self-consistency between values of basic nuclear parameters in the current database.[4] Five of six critical parameters for these three isotopes were found to be consistent[5] and the sixth was repaired. Lest this give a false sense of security in the use of the existing database for the description of other laser-related processes, it must be recognized that these three materials were chosen because it appeared <u>a priori</u> that they were the materials most precisely character-

ized by the study of particle reactions reported in the literature. The resulting score of 83.3% for consistency should be considered representative of only the very best group of test nuclei. Fortunately, the level of consistency was this high or XAN could not have been validated in a practical number of test shots.

The next logical step would be to add more isotopes to the calibration target in order to improve both resolution and coverage and a number of candidates suggest themselves. During the same series of experiments on the PITHON nuclear simulator during which self-consistency of the basic three was demonstrated, 29 other isotopes were irradiated in a survey mode. Four, ^{167}Er, ^{179}Hf, ^{191}Ir, and ^{197}Au, were found to be particularly promising and others were certainly interesting. The opportunity for using those four to add four more energies to the list for which XAN is applicable is reported in this manuscript. Unfortunately, of the ten nuclear parameters of importance *at least five accepted values were found to be drastically erroneous*. Such an error rate in excess of 50% for this part of the nuclear database, coupled with the lack of foreknowledge of which of the materials should have borne greatest emphasis, insured that the resulting data would be insufficient for the self-consistent extraction of the parameters needed for the application of XAN at four additional energies. Nevertheless, partial analysis of the data for these four species is both interesting and provocative. It is reported here and serves as a convenient point of departure for any subsequent effort designed to concentrate additional attention upon the definitive measurement of the "optical-like" parameters of these materials in the 0.1 - 1.5 MeV range of gamma-ray energies.

Method

In a previous report[1,2] it was shown that the uncertainty in the absolute value of the geometric coefficient coupling the source of pump radiation to the absorbing target could be eliminated by normalizing both the pump fluence and the fluorescence counts to some standard material having a monochromatic excitation spectrum. The reaction $^{79}Br(761,\gamma')^{79m}Br$ was found to be an ideal standard, having an integrated cross section of 6.2×10^{-29} cm^2 keV. Following the formalism reported earlier, the number of isomeric nuclei, S(x), of material x which could

be excited by a flash of intense bremsstrahlung can be conveniently expressed[1] as a ratio,

$$R(\text{model}) = \left.\frac{S(x)/N(x)}{S(Br)/N(Br)}\right|_{\text{Model}} = \sum_E \frac{\xi_E(x)}{\xi_{761}(Br)}\,\zeta(E) \quad , \qquad (1)$$

where S(x) and N(x) are the number of nuclei produced and the number of target nuclei of material x, respectively; $\zeta(E)$ is the ratio of pumping intensity at E keV to the intensity at 761 keV and the $\xi_E(x)$ are the combinations of nuclear parameters involved in the excitation of the gateway level at energy, E,

$$\xi_E(x) = \frac{(\pi b_a b_o \Gamma \sigma_o/2)_E}{E} \quad . \qquad (2)$$

The collection of terms in parenthesis in Eq. (2) comprises the integrated cross section for excitation as usually reported. They are summarized in Table I for the nuclei whose energy levels are found in Figs. 1 and 2.

Table I

Summary of literature values for nuclear parameters important to the excitation of isomeric fluorescence. These were not confirmed by the results of this experiment.

Nuclide	Ref.	E_{out} (keV)	E_{in} (keV)	$\pi b_a b_o \Gamma \sigma_o / 2$ ($\times 10^{-29}$ cm^2 keV)
^{167}Er	a	207	532	0.074
			667	0.036
			745	0.49
^{179}Hf	b	214*	650	0.057
			880	0.082
			1030	0.25
			1160	0.22
			1400	2.0
^{191}Ir	Ref. 4	129**	659	0.037
^{197}Au	Ref. 4	279***	None	

*cascading from the isomer at 375 keV
**cascading from the isomer at 171 keV
***cascading from the isomer at 409 keV

a. J. A. Anderson and C. B. Collins, Proof of the Feasibility of Coherent and Incoherent Schemes for Pumping a Gamma-Ray Laser, University of Texas at Dallas, Report #GRL/8602, Innovative Science and Technology Directorate of Strategic Defense Initiative Organization, April 1987, p. 45.

b. E. A. Henry, Nuc. Data Sheets 17, 287 (1976). These are average or effective levels and do not correspond to individual levels in Fig. 2.

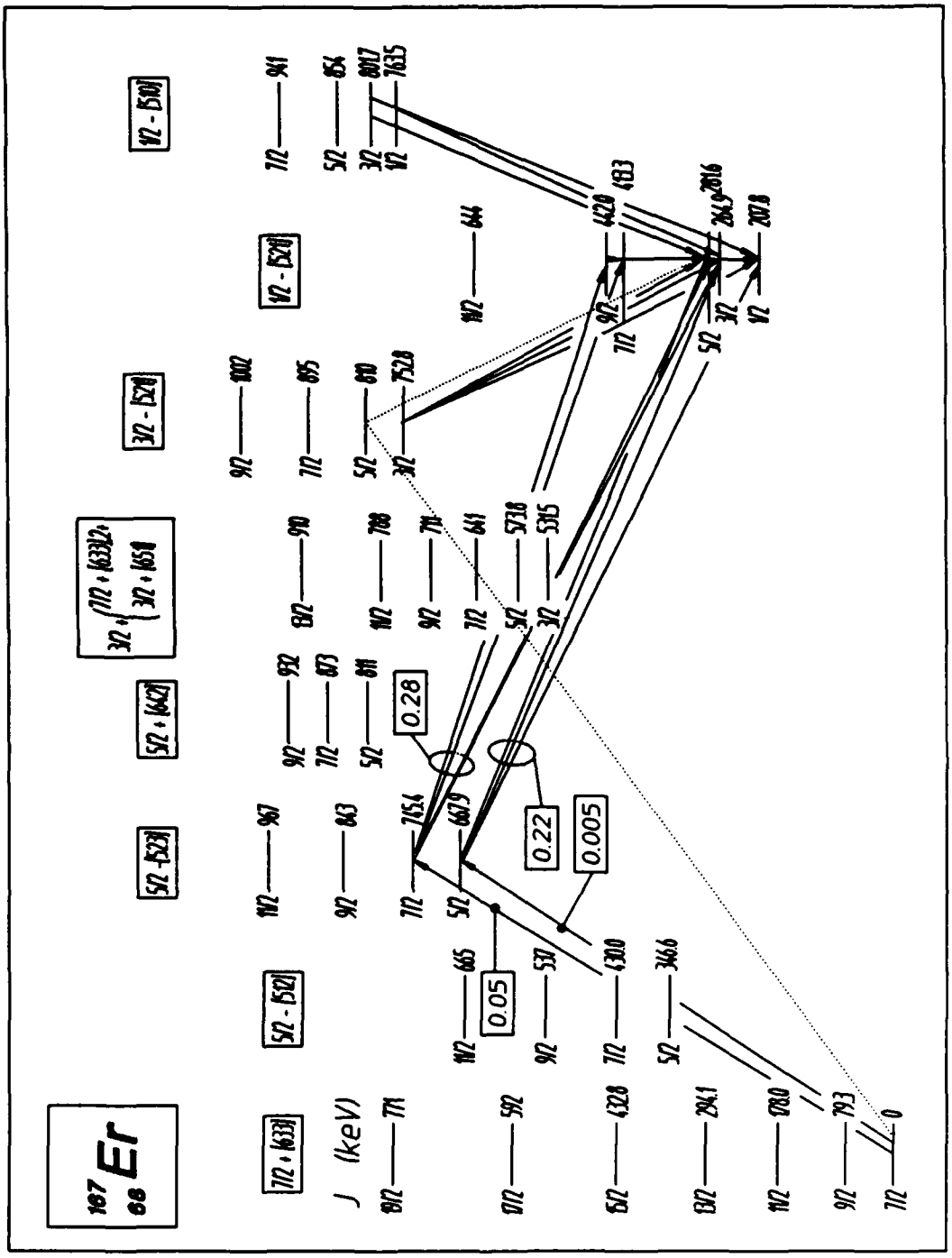

Figure 1: Energy level diagram of the spheroidal nucleus ^{167}Er showing a band structure reminiscent of the energy levels of diatomic molecules. Each column shows the ladder of rotational levels built upon the single particle state whose Nilsson quantum numbers given by Ref. 4 are shown in the box at the top of that column. Transitions to the isomeric state that are listed in Ref. 4 are shown by the arrows. Branching ratios are shown in boxes for two principal transitions for the (γ,γ') channels important in this work.

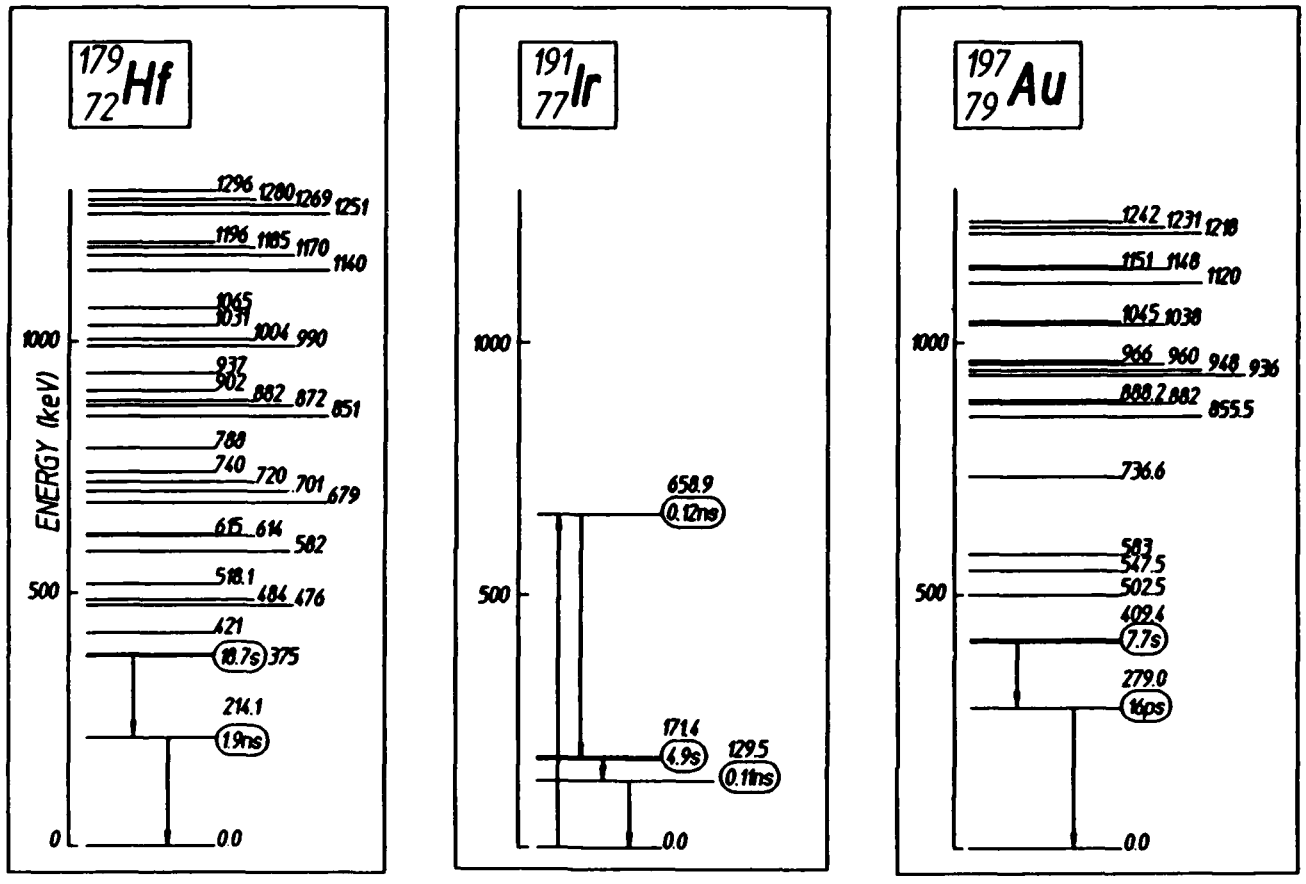

Figure 2: Energy level diagrams of the excited states of these nuclei that might be pumped from their ground states to cascade to their respective isomers. Arrows show paths of known (γ,γ') reactions.

The source of excitation in these experiments was the bremsstrahlung produced by the PITHON nuclear simulator at Physics International. The nominal end point energy of the electrons producing the bremsstrahlung was 1.3 MeV with relatively small shot-to-shot variance. For these particular experiments the nominal firing parameters were deliberately perturbed so that successive irradiations could be obtained with end point energies varying from 0.9 to 1.5 MeV.

Intensities at the target were determined by measuring the nuclear activation of the ^{79}Br component of a sample of LiBr containing isotopes in natural abundance. This calibrating target was run in a pneumatic transfer system which enabled the population of ^{79m}Br produced by a single irradiation to be subsequently counted at a quiet location 30 m removed from the source. Activation lost during the 1.0 sec transit time could be readily corrected during analysis.

The sample material, x, was placed in a second pneumatic shuttle and transferred after irradiation to another NaI(Tl) spectrometer system. Electronics were capable of simultaneously producing a record of counting rate as a function of time together with the spectrum as a function of energy. In that way it was assured that the lifetime of the activation was consistent with the literature value.

Standard procedures were employed for determining the efficiencies for the detector, for the emission of gamma fluorescence by the isomer and for the escape of radiation from the sample in the particular geometry employed. Correction was also made for the loss of activity during the individual transit time of the sample recorded after each irradiation.

Results

Because of a physical displacement of the shuttle system for the test material from the mixed ^{79}Br/^{77}Se target providing calibration, the actual number of fluorescent photons counted from the former had to be corrected for the extra path length from the source point to the absorber. This was done by mounting thermoluminescent diodes (TLD's) at both positions and then comparing the total dose recorded at the different points for each shot. The number of photons from the test

materials were scaled by the value of relative dose received at the test shuttle and at the calibrating positions.

The Hf and Au were in the form of foils mounted at the midplane of the cylindrical target shuttle. They were manually positioned for normal incidence before irradiation. Received after transfer into a well type geometry, no correction was necessary for angular orientation.

ERBIUM

The sample of erbium run in the test shuttle consisted of 2.76 g of Er_2O_3 containing natural isotopic abundances. The fluorescence spectrum is shown in Fig. 3, together with the result expected from Eq. (1) multiplied by the number of excitations observed in the bromine calibration and by the relative efficiencies. As can be seen, the experimental results exceed expectations by a large factor which computes to 17.

From Fig. 3 it can be immediately concluded that either one or more of the parameters for ^{167}Er listed in Table I is grossly incorrect or there is an excitation channel above 745 keV which is substantially more effective than those listed. Figure 1 shows a high density of states which might seem to offer numerous possibilities. However, ^{167}Er is the most elongated of the stable nuclei and hence the one most nearly conforming to the Nilsson model of structure.[6] The energy level diagram of Fig. 1 is dominated by rotational bands built upon an initial value of K, the quantum number for the projection of nucleonic angular momentum upon the axis of elongation. The situation is very analogous to that for diatomic molecules.

Transitions must not only satisfy selection rules upon changes in J, the total angular momentum, but also upon K; both ΔJ and ΔK being limited by the multipolarity of the transition. The limitations upon ΔK are understood to comprise the punishing "K-selection rules" which would seem to impede some of the otherwise attractive systems for a gamma-ray laser. Laser candidates suffering a large ΔK are currently excluded from serious consideration, automatically. Now, here is a K-hindered analog where fluorescence yield is 17 times expectations.

The change in K from ground to isomer in ^{167}Er is a negative three. It is not clear that there could be a reasonable path containing three steps for which $\Delta K = -1$ and $\Delta J = \pm 1$ or 0. Failing that, it is not clear how there could be a two step (γ,γ') reaction through which $\Delta K = -2$ for

one component that would be any more probable than the known[4] E2 and M2 channels shown in Fig. 1. Those give the result which is too small by a factor of seventeen. Of course, there could be a more favorable E2 route if transitions to one of the states were enhanced by a collective quadrupole vibration, but the one band for which this is predicted is not reported[4] to be radiatively connected to ground and isomeric states. *Whatever the reason, the experimental fact is that an excitation sequence exists which has the effect of short-circuiting the K-selection rules as usually applied in excluding low priority candidates for a gamma-ray laser.*

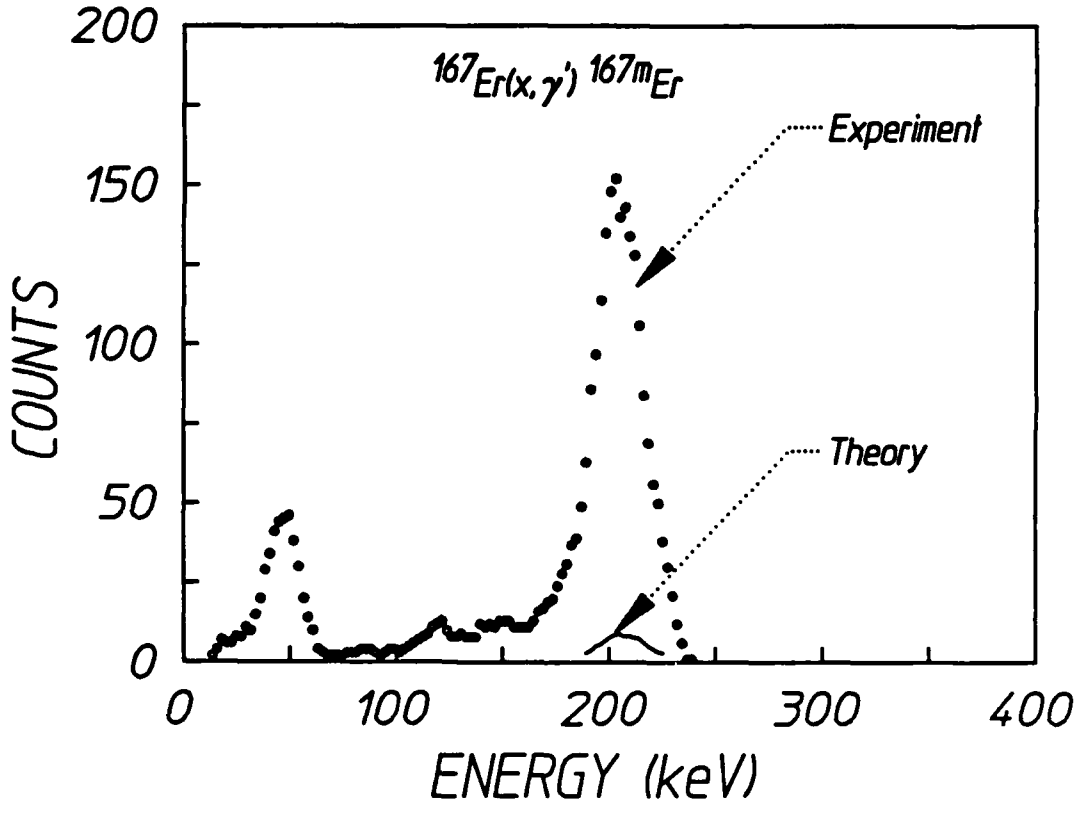

Figure 3: Arrows identify the nuclear component of the spectrum of fluorescence of Er_2O_3 in comparison with model predictions based upon nuclear parameters input from the extant database. Data were obtained from a single irradiation of 2.8 g of Er_2O_3 in natural isotopic abundance.

The particularly effective (γ,γ') sequence in ^{167}Er can be identified through the same iterative procedures used to discover and characterize the dominant $^{77}Se(1005\ keV, \gamma')^{77m}Se$ channel in the presence of

the three other known reactions producing the same product.[1,2] In implementation it requires a sequence of measurements of fluorescence yield be made with successively differing values of end point energies of the bremsstrahlung. Because of the survey nature of the series reported here, only a single, fully instrumented measurement was obtained and so the analysis which can be presented at this time is limited to the single conclusion that there is a major channel for (γ,γ') reactions in ^{167}Er which should provide another useful energy at which XAN techniques can be applied.

HAFNIUM

As can be seen from Table I, ^{179}Hf is an isotope for which the spectrum for activation is even more complex than ^{167}Er or ^{77}Se. Having five distinct (γ,γ') reactions for the production of isomers, it places even greater demands upon the self-consistency of the five critical nuclear parameters in Table I. Moreover, it is particularly curious that the transition energies for the gamma steps in Table I correspond to no individual levels in the scheme of Fig. 2, currently accepted.[4] They evidently represent the sums of excitations through gateway levels whose average positions in Fig. 2 would lie at the energies listed in Table I. It is of considerable interest that no absorption transitions from the ground state in the energy range 375 - 1500 keV are listed in accepted data bases.[4] From this perspective it is almost surprising that experimental results approached the model predictions of Eq. (1) to within the factor of 2.0 - 2.7 actually observed.

The fluorescence spectrum obtained from a single irradiation of 4.71 g foil of hafnium in natural isotopic abundance is shown in Fig. 4. Also shown is the comparative value obtained from Eq. (1) using the parameters of Table I placed into the same units of detected counts by multiplying Eq. (1) by the number of ^{79}Br excitations times the relative efficiencies for fluorescence, escape and detection. While indicating that one or more of the parameters for ^{179}Hf in Table I are underestimating the actual value of integrated cross section, the suggested discrepancy of 2.0 - 2.7 is more of the magnitude which might be reasonably expected to result from a direct remeasurement of such derived parameters.

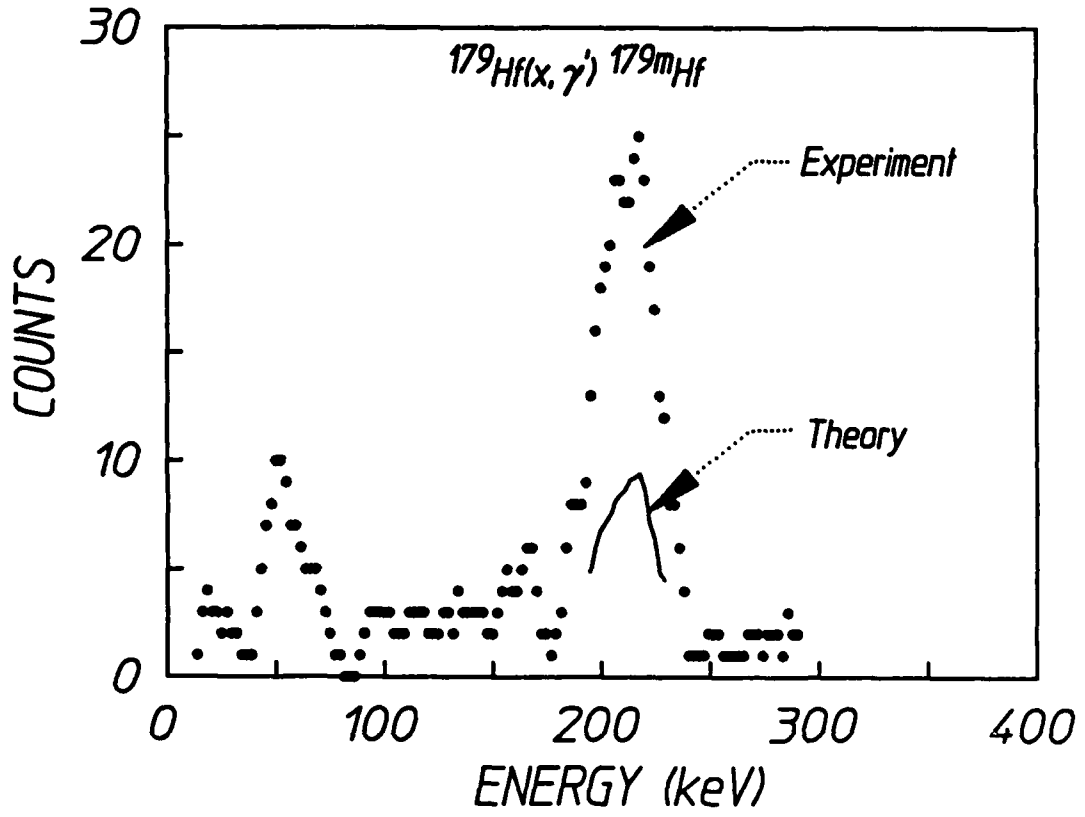

Figure 4: Arrows identify the nuclear component of the spectrum of fluorescence of Hf in comparison with model predictions based upon nuclear parameters input from the extant database. Points were obtained from a single irradiation of 4.71 g of metal in natural isotopic abundance.

As in the case of ^{77}Se, attempts to identify the extra contribution should begin with a computation of the residue, Δ, between the measured ratios of the numbers of ^{179m}Hf to ^{79m}Br,

$$\Delta = R(\text{exp}) - R(\text{model}) \quad , \qquad (3)$$

where the R is as defined in Eq. (1). This was not done in this case because R(exp) >> R(model) so that the observed ratio could be reasonably expected to show the same correlations with experimental variables as would the residue Δ.

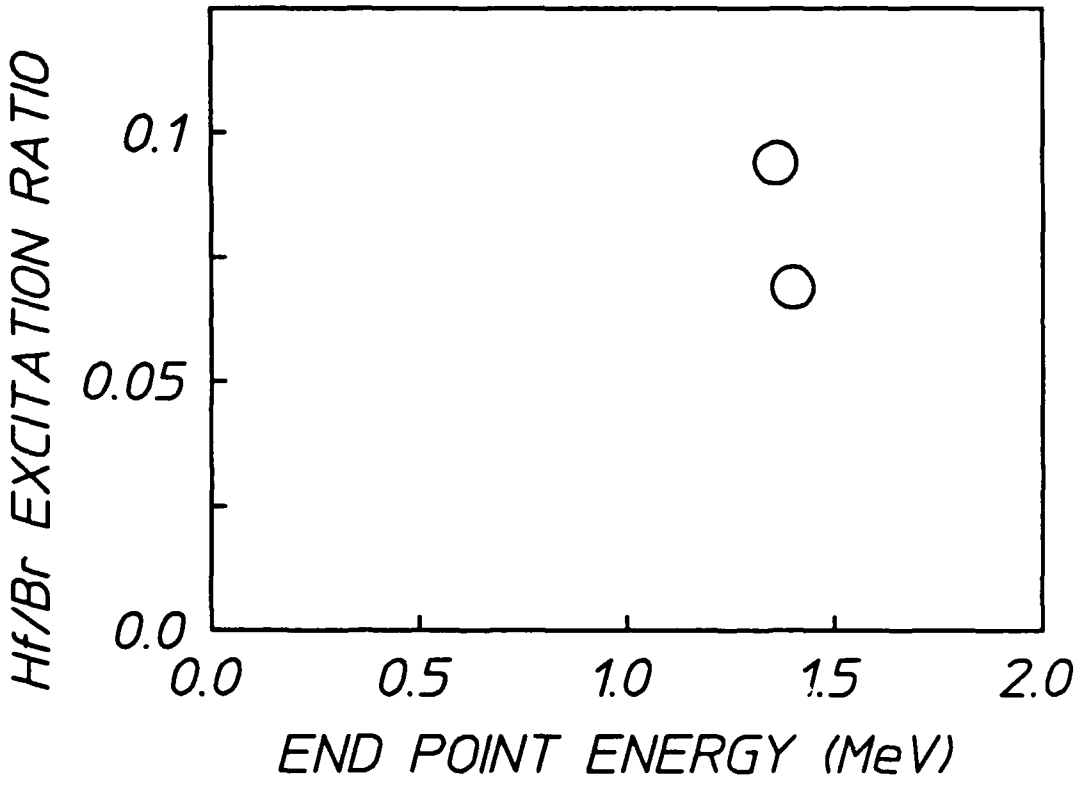

Figure 5: Relative numbers of isomers ^{179m}Hf and ^{79m}Br normalized to equal numbers of initial targets plotted as functions of the end point energy of the electron beam producing the bremsstrahlung.

Figure 5 plots the resulting data[7] for the only two observations fully instrumented as functions of the end point energy of the bremsstrahlung. While statistical uncertainty is less than the size of the plotted points, the trend in Fig. 5 is not considered significant. As in the previous example, here also a larger number of data points is needed to identify and characterize the dominant reaction.

IRIDIUM

The most drastic inconsistency in this group was found for ^{191}Ir, which should have the simplest scheme for excitation. The spectrum of (γ,γ') reactions leading to the production of the ^{191m}Ir isomer is reported to be monoenergetic,[4] as shown in Fig. 2. If so, this would place a very high importance on ^{191}Ir as a material contributing another clearly selective line for sampling energies in the XAN procedure.

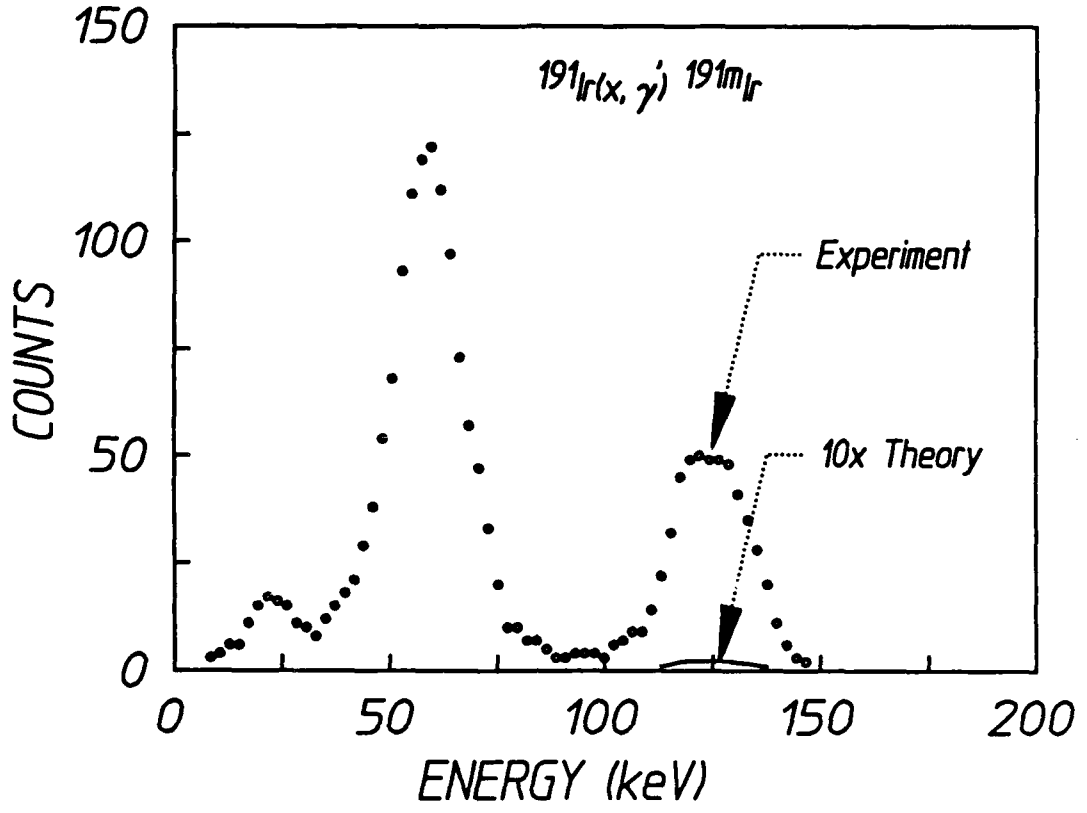

Figure 6: Arrows identify the nuclear component of the spectrum of fluorescence of Ir in comparison with model predictions based upon nuclear parameters input from the extant database. Points were obtained from a single irradiation of 1.82 g of metal in natural isotopic abundance.

The iridium sample used in these experiments had the form of 1.82 g of metallic powder in natural isotopic abundance sealed into a thin rectangular packet. The fluorescence spectrum is shown in Fig. 6, and its time decay positively identified the source as ^{191m}Ir. Self-absorption of the fluorescence at 129 keV represented a significant effect in this geometry and data were corrected appropriately. In the units of Fig. 5 and the left side of Eq. (1), the measured value in the peak was

$$R(exp) = 1.86 \quad , \qquad (4)$$

in comparison to the values near 0.1 encountered in the previous example. This immediately identifies ^{191}Ir as possessing a path for (γ,γ') reactions of major importance.

If it is assumed that the reaction proceeds through the single reactive channel, ^{191}Ir(659 keV, γ')^{191m}Ir, as suggested in the literature, the right-hand side of Eq. (1) consists of a single term when Eq. (4) is substituted,[7]

$$R(exp) = \frac{\xi_{658}(Ir)}{\xi_{761}(Br)} \zeta(658) \quad . \tag{5}$$

Substituting[8] and solving for ξ_{658}(Ir),

$$\xi_{658}(Ir) = 12.6 \times 10^{-32} \text{ cm}^2 \quad . \tag{6}$$

Thus, from Eq. (2),

$$\pi b_a b_o \Gamma \sigma_o/2 = 8.3 \times 10^{-29} \text{ cm}^2 \text{ keV} \quad , \tag{7}$$

meaning that the half life of the funneling level at 658 keV is 0.6 ps rather than the value of 120 ps generally accepted.[4]

An alternative explanation is that there is a previously unobserved channel at higher energies, as was the case in ^{77}Se. As an example, if we arbitrarily suppose that the reaction proceeds through ^{191}Ir(1000 keV, γ')^{191m}Ir then the indices, 658 in Eq. (5) are replaced by 1000. However ζ(1000) is much smaller than ζ(658) so the solution of Eq. (5) in that case for Eq. (4) would give an even larger integrated cross section of

$$\pi b_a b_o \Gamma \sigma_o/2 \sim 60 \times 10^{-29} \text{ cm}^2 \text{ keV} \quad , \tag{8}$$

a very large value for a total spin change of $\Delta J = -4$ between ground and isomeric states.

Either way, whether there is a shockingly broad funneling state for such an unfavorable spin change, or whether the accepted values[4] of more modest transitions are in error by a factor of over 200, the ^{191}Ir isotope spotlights itself as an important candidate for a more extensive investigation.

GOLD

The system of (γ,γ') channels for the production of isomeric gold nuclei is the simplest possible--there are no channels.[4] Nevertheless, samples of gold activate quite strongly, at a qualitative level comparable to ^{179}Hf.

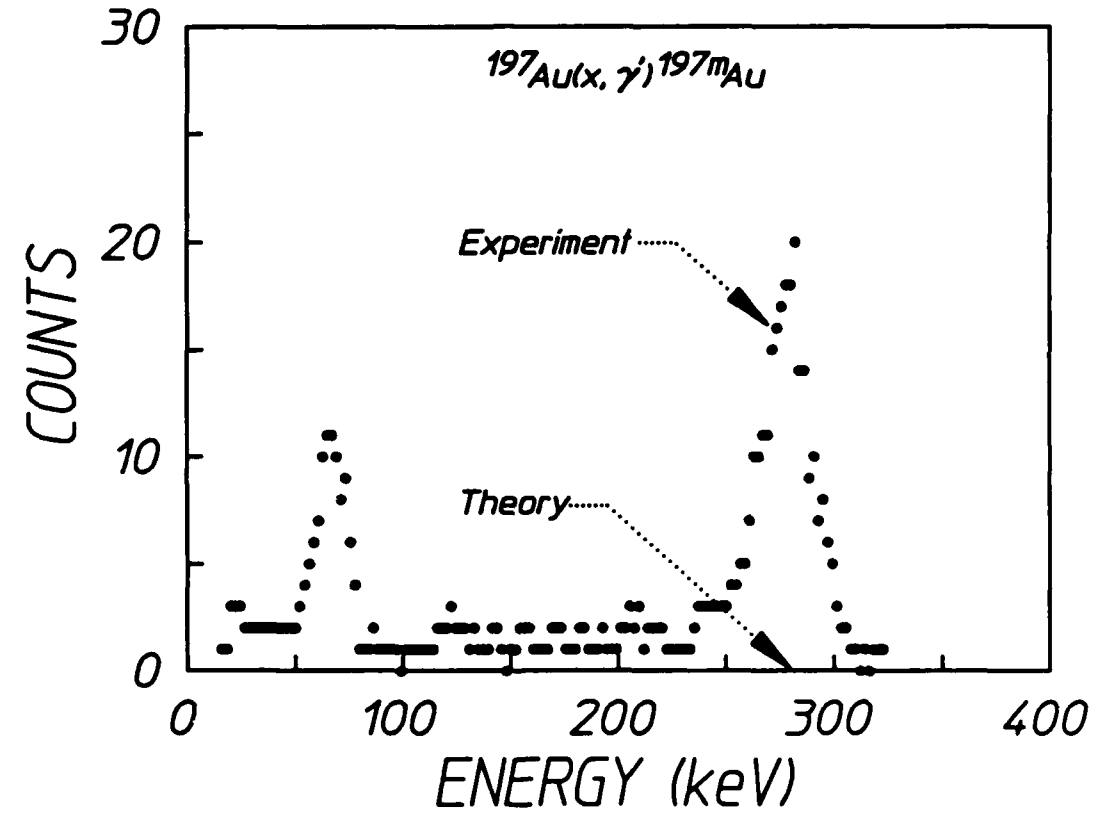

Figure 7: Arrows identify the nuclear component of the spectrum of fluorescence of Au in comparison with model predictions based upon nuclear parameters input from the extant database. Points were obtained from a single irradiation of 2.41 g of metal in natural isotopic abundance.

In this survey a gold foil of 2.41 g was mounted in a shuttle target with the same geometry as characterized the hafnium experiments. Commercial gold foil was used in which the ^{197}Au isotope occurs with natural 100% abundance. The spectrum of a single irradiation is shown in Fig. 7 and identification was confirmed by the time dependence of the fluorescence. As in previous examples, data were reduced to ratios of

the numbers of isomeric nuclei per unit concentration and the level of excitation on this scale was of the order of 0.1, as with hafnium.

Two irradiations were obtained under almost ideally contrasting values of end point energies for the bremsstrahlung. Experimental values of R from Eq. (1) are plotted in Fig. 8 for the two available shots. Statistical uncertainty of counting is much smaller than the size plotted for the data points.

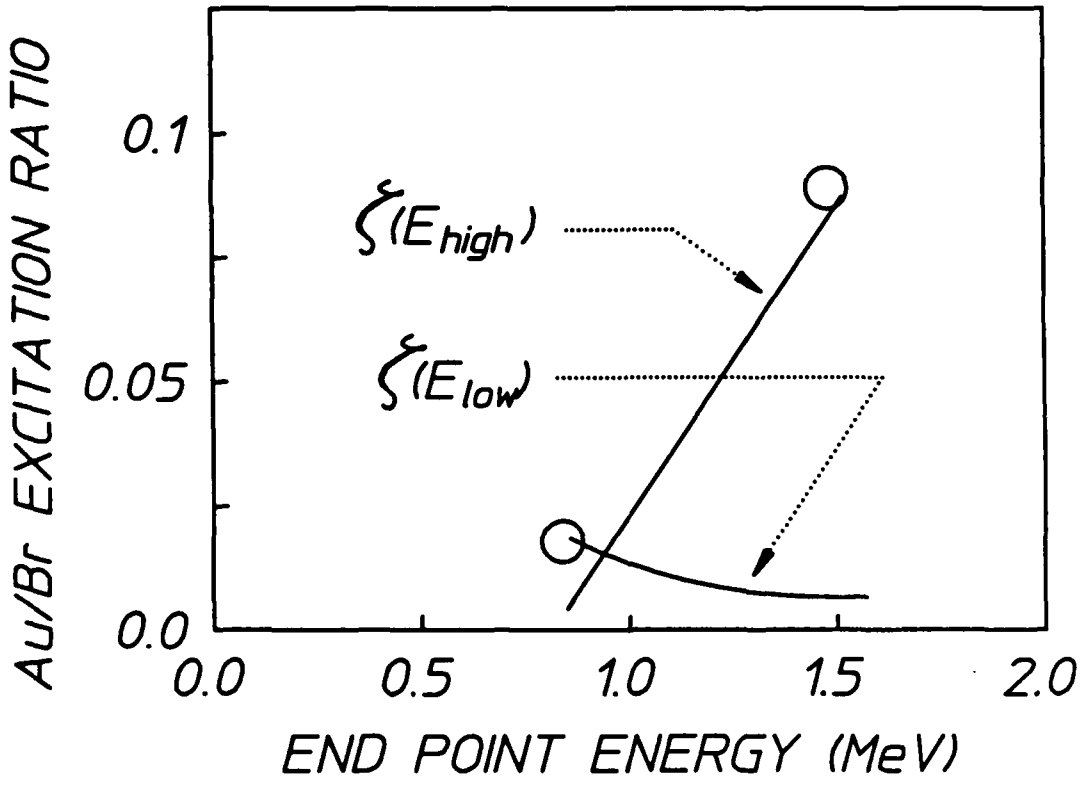

Figure 8: Relative numbers of isomers ^{197m}Au and ^{79m}Br normalized to equal numbers of initial targets plotted as functions of the end point energy of the bremsstrahlung. Curves show the variations of the intensities at high and low pump energies relative to the intensity pumping the ^{79m}Br.

As is readily seen from Eq. (5), if a single reactive channel is dominant the dependence of the data points upon end point energy should correlate with the variations of the relative intensity, $\zeta(E)$, where E is the energy of the gateway state. The shapes of curves of $\zeta(E)$ as functions of end point energy differ drastically[2] for different excitation energies, E. As in the case[1,2] of ^{77}Se, a simple visual comparison

of data with the different shapes[2] can give a clue to the value of the excitation energy, E, of the gateway state. Of course, a substantial number of data points is needed for such comparisons to be unequivocal.

Shown in Fig. 8 are two typical curves of $\zeta(E)$, one for a high value of excitation energy and one for a lower value. For the higher values they tend to "point" to an intercept near the gateway energy.

In the case of Fig. 8, it is tempting to identify the gateway as occurring in the cluster of levels near 900 keV. However, also as suggested by the placement of the curve of $\zeta(E)$ for a lower energy threshold, a contribution from a second gateway at lower energy could displace the apparent intercept to lower values giving a quite erroneous conclusion were a system with two gateways analyzed as having only one. Nevertheless, it is interesting to estimate the magnitude of the integrated cross section for excitation were it to arise from a single level in ^{197}Au near 900 - 1000 keV. In that case $\zeta(1000) \sim 0.26$ for an end point energy near 1.4 MeV. Then from the analog to Eq. (5),

$$\xi 1000(\mathrm{Au}) \sim 2.8 \times 10^{-32}\ \mathrm{cm}^2 \quad , \tag{9}$$

and for ^{197}Au(1000 keV, γ')^{197m}Au,

$$\pi b_a b_o \Gamma \sigma_o / 2 \sim 2.8 \times 10^{-29}\ \mathrm{cm}^2\ \mathrm{keV} \quad , \tag{10}$$

provided excitation actually occurs through a single gateway in the 900 - 1000 keV range of energies.

Conclusions

Attempts, a posteriori to extract new lines for the XAN procedure for calibrating intense pulses of bremsstrahlung from the survey data were as frustrated as they should have been. Not nearly enough data were available for the sort of painstaking analyses of residues proven effective in the reconciliation of the nuclear data for ^{77}Se. However, some provocative indications were found.

Whereas existing values of nuclear parameters were found to be 83% self-consistent in the best set of isotopes, ^{77}Se, ^{79}Br and ^{115}In; in this new group of four, ^{167}Er, ^{179}Hf, ^{191}Ir and ^{197}Au, extant parameters were consistent in fewer than 50% of the cases. Moreover, discrepancies were

drastic. Fluorescence from ^{167}Er should have been hindered by the same punative K-selection rules which have been used to downgrade priorities of some laser candidates; but actual measurement showed 17 times more yield than expected. Experimental excitation evidently finds a real path around the K-rules which should have been the most restrictive for this ^{167}Er nuclelus because that is the stable isotope most nearly conforming to input assumptions to the theory. It is the most elongated stable nucleus.

Fluorescence from ^{179}Hf was 2.0 to 2.7 times more than predicted from seemingly complete data spanning the range of energies up to 1.4 MeV. Negligible within the context of the more extreme discrepancies found in this group of four, it is an unaccountable excess far greater than was observed in ^{77}Se, the worst example in the previous group studied.

In ^{191}Ir the largest finite excess of experiment over theory was found. Either representing an error of 20,000% in accepted values or the dominance of a new gateway of tremendous width, the results with this isotope merit description as being shockingly unexpected. Such results in this group of four offer an extreme contrast to the smooth agreement obtained in the first group of isotopes with 83% of the important parameters being found to be correct.

For ^{197}Au the results described in the work came the closest to providing insight into the discrepancy which is becoming the majority result--experimental excitation of isomeric fluorescence is much more effective than indicated by the "accepted" database. This case benefited from the largest variation of input spectra and so suggested a trend. Far from compelling belief, nevertheless the two data points for ^{197}Au suggest a threshold behavior. If so, the gateway state would be one of the cluster in the 900 - 1000 keV range and would support an integrated cross section for funneling to the isomer of 2.8 ($\times 10^{-29}$ cm^2 keV) units in the system generally employed. This is a surprisingly large value to find so easily for a completely unexpected pumping sequence spanning a change of angular momentum of $\Delta J = 4$. In a sense, for gold experimental yield is infinitely greater than theory because accepted models[4] provide no channels for such an excitation.

Such results as presented here spotlight the drastic inadequacies of the existing database for the purposes of screening candidates for a

gamma-ray laser. These earliest results cast severe doubt onto the validity of generally accepted procedures invoking K-selection rules for reducing the slate of candidates by elimination. *It appears that instead of narrowing the field of candidate laser isotopes, these experiments are widening the field.* Much more study is needed to see whether the experimental production of yields from heavy nuclei that are drastically in excess of theory will prove to be a general phenomenon.

References

1. J. A. Anderson and C. B. Collins, Rev. Sci. Instrum. (II pending).

2. J. A. Anderson and C. B. Collins, Proof of the Feasibility of Coherent and Incoherent Schemes for Pumping a Gamma-Ray Laser, University of Texas at Dallas, Report #GRL/8701, Innovative Science and Technology Directorate of Strategic Defense Initiative Organization, July 1987, pp. 11-34.

3. C. B. Collins, F. W. Lee, D. M. Shemwell, B. D. DePaola, S. Olariu, and I. I. Popescu, J. Appl. Phys. 53, 4645 (1982).

4. Evaluated Nuclear Structure Data File (Brookhaven National Laboratory, Upton, New York, 1986).

5. Table I of Ref. 2.

6. A. deSchalit and H. Feshback, Theoretical Nuclear Physics Vol. I: Nuclear Structure (J. Wiley, New York, 1974) Ch. 6, Section 14.

7. In contrast to figures used in our recent descriptions of the activations of ^{77}Se, ^{79}Br and ^{115}In, these data are the ratios of the number of isomeric nuclei produced in the targets per constant number of original ground state nuclei and need not be corrected further before use in Eq. (1).

8. From Ref. 1, $\xi_{761}(\mathrm{Br}) = 8.15 \times 10^{-32}$ cm^2 and $\zeta(658) = 1.2$.

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